

^{237}Np , ^{238}Np , ^{235}U , ^{233}U , and ^{239}Pu ;

FISSION YIELD STUDIES

^{237}Np , ^{238}Np , ^{235}U , ^{233}U , AND ^{239}Pu ;

FISSION YIELD STUDIES

By

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A Thesis

Submitted to the Faculty of Graduate Studies

in Partial Fulfilment of the Requirements

for the Degree

Doctor of Philosophy

McMaster University

May 1969

DOCTOR OF PHILOSOPHY (1969)
(Physics)

McMASTER UNIVERSITY
Hamilton, Ontario.

TITLE: ^{237}Np , ^{238}Np , ^{235}U , ^{233}U , and ^{239}Pu ;
Fission Yield Studies.

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SUPERVISOR: Professor H. G. Thode

NUMBER OF PAGES: x, 143

SCOPE AND CONTENTS:

The relative cumulative yields of krypton and xenon isotopes from the thermal neutron fission of ^{237}Np and ^{238}Np and from the fast neutron fission of ^{237}Np have been measured by means of a mass spectrometer. These are the first fission yield results for ^{238}Np , and the first for ^{237}Np at thermal neutron energies. The results are compared with those from other fissioning nuclides.

Independent yields of the shielded nuclides ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I from the thermal neutron fission of ^{235}U , ^{233}U , ^{239}Pu , and ^{238}Np have been determined by mass spectrometric analyses of the krypton and xenon β -decay products. The results are discussed in terms of conventional charge distribution theories, and also in terms of neutron emission effects.

ACKNOWLEDGEMENTS

I am deeply indebted to my supervisor, Dr. H. G. Thode, for his encouragement and guidance throughout the course of this investigation. I also wish to express my appreciation to Dr. R. H. Tomlinson, Dr. W. B. Clarke, and Dr. J. F. Pleva for many valuable discussions.

Dr. T. A. Eastwood of Atomic Energy of Canada has kindly assisted in carrying out the irradiations at the Chalk River Nuclear Laboratories. Mr. Gary Kugler has provided the ^{235}U samples which were purified by induction heating. A word of appreciation is also in order for Mrs. Stella Smith, who typed this thesis, and for my wife, Dianne, who drew the illustrations.

Finally, I am grateful to the National Research Council of Canada for generous financial assistance from 1964 to 1968.

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INTRODUCTION

NUCLEAR FISSION

In the three decades that have elapsed since Otto Hahn and Fritz Strassman (1) discovered nuclear fission in 1939, the world has seen a virtual explosion in the development of nuclear science and technology. Nuclear weapons and atomic power generation are now realities. The nuclear reactor has proven to be a most valuable tool for the study of nuclear reactions and crystal structures, for the synthesis of new elements, and for the production of a host of useful radioisotopes. Yet the phenomenon of nuclear fission itself is still not completely understood.

Nuclear fission is the splitting of a heavy nucleus, such as ^{238}U , into two medium-weight fragments plus several free neutrons. This process releases a large amount of energy--about 200 Mev per fission. The free neutrons can initiate further fissions, and with highly fissionable nuclides such as ^{235}U , ^{233}U , or ^{239}Pu , a self-sustaining reaction can be set up providing a tremendous output of power from a relatively small quantity of fuel. Fission is known to occur, to a greater or lesser extent, in most of the heavy elements. It can be induced by bombardment with neutrons or high energy charged particles, or it can occur spontaneously, such as in ^{238}U or ^{252}Cf , by means of barrier penetration. Insight into the fission process

can be gained by detecting the various fission products and measuring their frequencies of occurrence, kinetic energies, and other properties. This thesis describes measurements of cumulative krypton and xenon yields from the fission of neptunium. This gives information on the distribution of mass in fission. Measurements of independent bromine and iodine yields from a number of fissioning species are also described. This is related to the problem of nuclear charge distribution in fission.

In order to clarify some of these terms, let us consider the history of the massive fission fragments after their final separation in fission. The two fragments immediately repel one another and attain high kinetic energies. Being a highly excited state, each fragment then emits one or two prompt neutrons followed by several γ -rays. Since the neutron-to-proton ratio of the fissioning nucleus is considerably higher than that of stable medium-weight nuclides, the fission fragments will generally have an excess of neutrons even after prompt neutron emission has occurred. They must still undergo several β -decays in order to reach stability. An example of this sequence of events is shown in Fig. 1. The newly-formed fragment, ^{133}Sn , first of all emits a prompt neutron and several γ -rays. The resulting nuclide, ^{132}Sn , then undergoes a succession of β -decays until the stable ^{132}Xe is formed. Such a series of isobaric nuclides is called a mass chain.

A fission product nuclide such as ^{132}I can be formed either

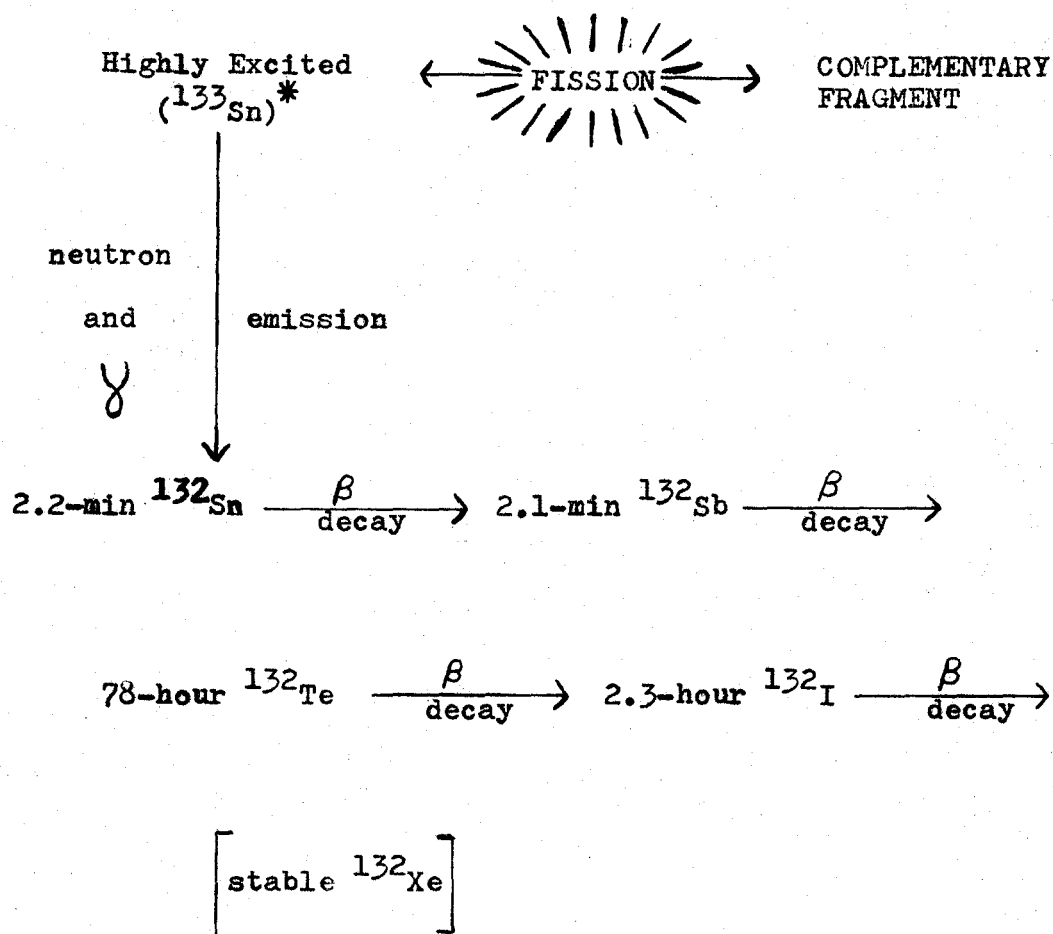


Fig. 1. A typical fission product mass chain.

directly from fission or indirectly from the β -decay of fission product precursors. The primary or independent yield of a certain nuclide is defined as the probability per fission, usually expressed as a percentage, that that particular nuclide will be formed directly in fission. This refers to the abundance of the nuclide after prompt neutron emission, but before any β -decays have occurred. The cumulative yield of a certain mass chain is the sum of all the independent yields along that chain. Expressed in another way, the cumulative yield refers to the probability of a given partition of nuclear mass, whereas the independent yields along a given mass chain refer to the various partitions of nuclear charge at that mass number.

For illustrations of the preceding definitions, consider the 132 mass chain in Fig. 1. After a few weeks, all the chain members will have decayed to stable ^{132}Xe . A measurement of the abundance of this nuclide then gives the cumulative yield at mass number 132. On the other hand, if the 2.3-hour ^{132}I is isolated before significant decay of the 78-hour ^{132}Te , then the measured abundance of ^{132}I gives the independent yield of that nuclide. This quantity divided by the cumulative yield at mass 132 gives the fraction of the 132 chain formed initially with nuclear charge (atomic number) 53.

A study of fission yields, then, divides itself naturally into considerations of cumulative yields (mass distribution) and independent yields (charge distribution).

CUMULATIVE YIELDS

Radiochemistry and the Mass Yield Curve

All the earlier fission yield data were obtained by radiochemical methods. Basically, the sample, which has undergone fission, is first dissolved. In order to recover the minute amounts of fission products, an inactive carrier of the particular element to be studied is added to the solution. Carrier and "tracer" together are then extracted by standard chemical techniques. The radioactive fission-product isotopes are identified and their abundances measured by radioactive counting.

With the development of high resolution counting equipment and refined techniques, radiochemistry is still a valuable tool in fission product studies. However, its overall accuracy is limited to about 10 percent because of uncertainties in half-lives and counting efficiencies, and also because of incomplete chemical separations. The scatter of mass yield data thus obtained is such that only a smooth curve can be drawn through the experimental points.

The radiochemical cumulative yield curve from the thermal neutron fission of ^{235}U is shown in Fig. 2 (2). This illustrates the general asymmetric character of the mass distribution. The two peaks are centred about mass numbers 139 and 95, although yields have been observed all the way from mass 72 to 164. As the mass of

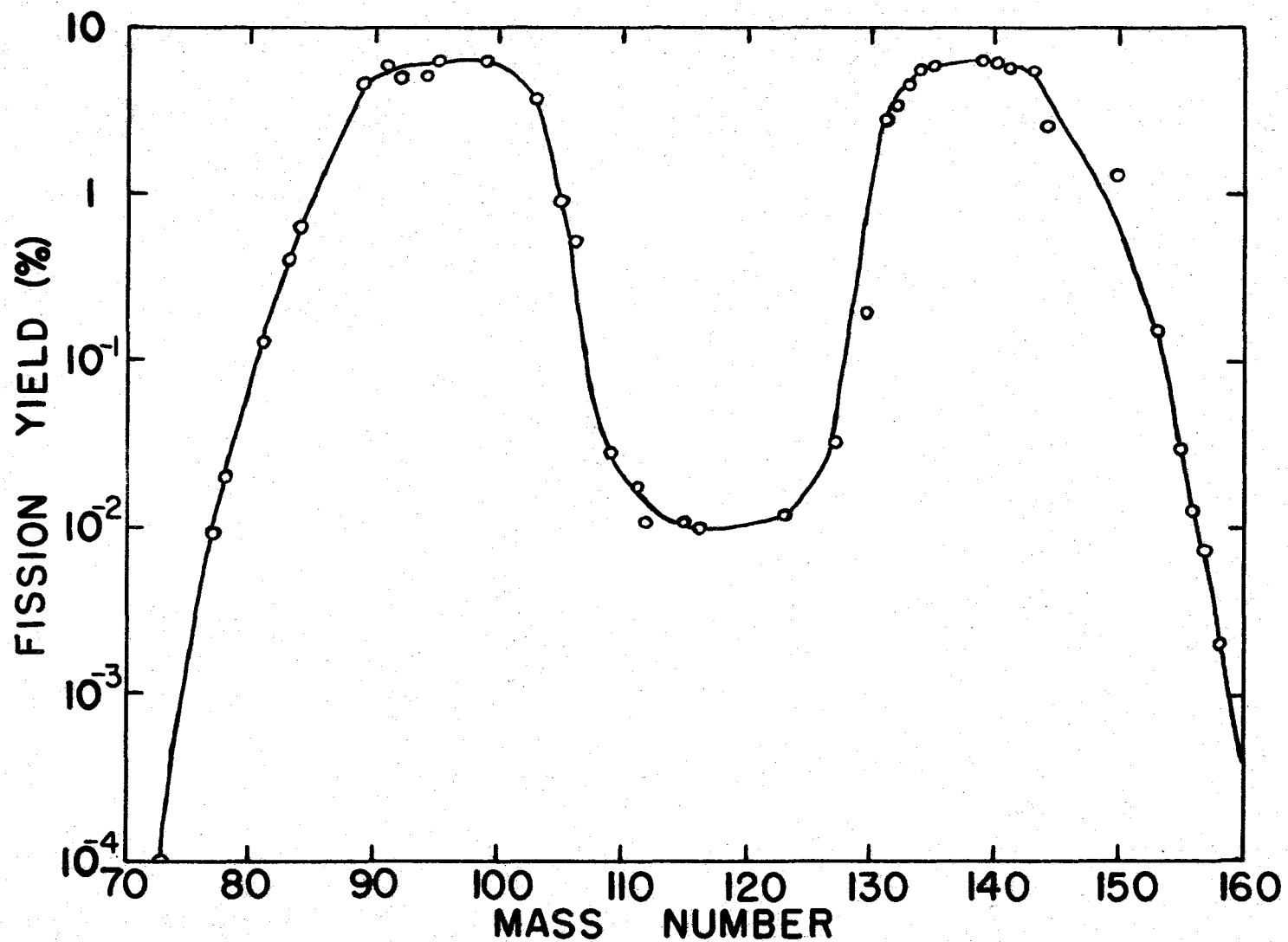


Fig. 2. Early radiochemical data(2) on the cumulative yields from the thermal neutron fission of ^{235}U .

the fissioning nuclide increases, the heavy mass peak remains more or less fixed in position while the light mass peak shifts to progressively higher mass numbers (3). The peak-to-valley ratio is taken as an indication of the relative occurrence of asymmetric versus symmetric splitting. In the case of thermal neutron fission of ^{235}U , this ratio is about 600 to 1. As the energy of the bombarding neutrons increase, symmetric fission becomes more probable. For 14 Mev neutrons, the peak-to-valley ratio is 6 to 1 (4).

The Mass Spectrometer and Fine Structure

The mass spectrometer offers a considerable improvement in the precision of fission yield determinations. It can measure the relative abundances of the isotopes of a certain element to within 0.1 percent. If this element is fission-product xenon, for example, then the 131, 132, 134 and 136 isotopes represent the stable end-products of four mass chains. Thus the relative yields at these mass numbers are obtained immediately. Absolute yields can be determined from the relative yields by using the techniques of isotope dilution or normalization.

The first mass spectrometric determinations of fission yields were carried out by Thode and Graham (6) at McMaster University. They analyzed the rare gases xenon and krypton produced in the thermal neutron fission of ^{235}U and obtained a ^{134}Xe yield that lay 35 percent above the smooth radiochemical mass yield curve. This fine structure at mass 134 has also been observed in the thermal neutron fission of

^{239}Pu (7), ^{241}Pu (8), ^{241}Am (9), and ^{242}Am (9), but not ^{233}U (10).

In the fission of ^{238}U (11) by fission spectrum neutrons, the fine structure appears to be shifted toward lower mass numbers. On the light mass peak, the yields of ^{84}Kr are abnormally high in most of the fission reactions studied (10, 11, 12, 13). There are, of course, instances of fine structure in other regions of the mass yield curve, but this thesis will be primarily concerned with yields in the 128-136 and 80-86 regions. Fig. 3 shows the complete mass yield curve for ^{235}U thermal neutron fission taken from recent mass spectrometric data (14).

Fine structure in the mass yield curve can result in two ways:

- (1) A preference in the fission act itself for fragments with certain closed-shell configurations would tend to produce abnormally high yields at some mass numbers.
- (2) A variation in the number of post-fission neutrons emitted from different fragments would give rise to local fluctuations in the cumulative yield curve.

Glendenin (15) made use of the second approach in an attempt to explain the high yields in the vicinity of mass 134. He suggested that fragments left with 83 neutrons after the usual prompt neutron emission would tend to evaporate an extra neutron in order to achieve an 82-neutron shell configuration. For instance, in ^{235}U fission, the most probable fragment normally produced at mass 135 is ^{135}Te (83-neutrons). In the Glendenin scheme, this nuclide would boil off

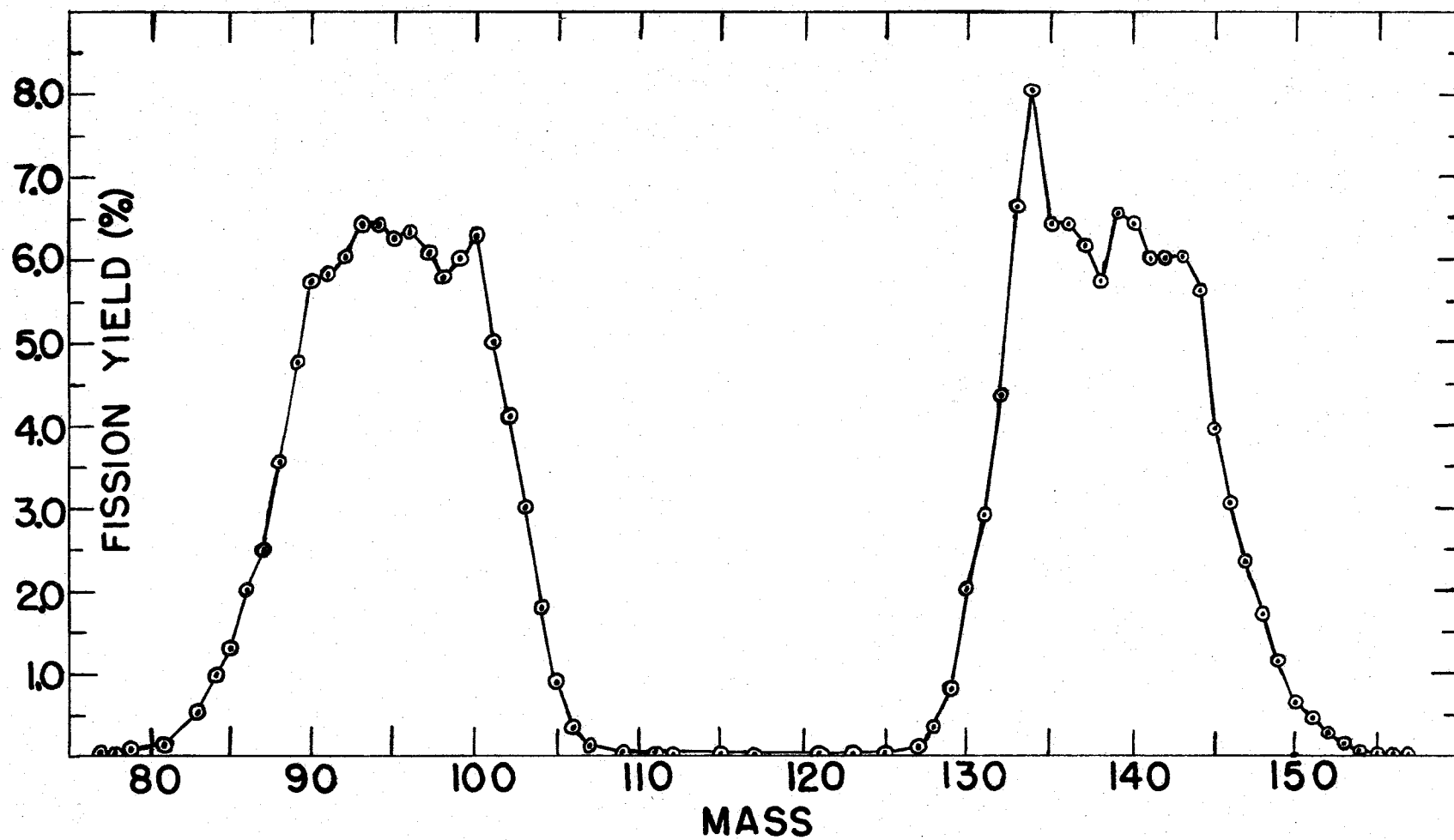


Fig. 3. Mass spectrometric data (14) on the cumulative yields from the thermal neutron fission of ^{235}U .

an extra neutron to enrich the 134 chain. On the other hand, the most probable fragment at mass 134, ^{134}Te (82 neutrons), would resist emitting an extra neutron. Thus the 134 yield would be greatly augmented. The difficulty with this hypothesis is that it predicts a depression of the 135 yield equal to the enhancement at mass 134. The observed 135 yield is not abnormally low.

Wiles et al. (16) proposed that the anomalously high yield at mass 134 results from a preference in the fission act itself for an 82-neutron shell configuration. The discovery of a similar high yield at the complementary mass number of 100 in ^{235}U fission (17) seems to substantiate this hypothesis. However, in ^{239}Pu fission, the yields at mass numbers complementary to 134 do not lie above the smooth curve (18).

Farrar and Tomlinson (19) proposed another explanation of the fine structure after a careful study of the cumulative yield curve of ^{235}U fission. They concluded that the number of neutrons emitted per fission decreases sharply in going from mass 144 to mass 132. If this decrease in neutron emission were associated with the heavy fragment, then one would expect a piling up of yields in the 136-132 region. This explanation avoids the difficulty of the Glendenin mechanism by not requiring a depression of the observed yield at a higher mass number.

The Farrar-Tomlinson explanation receives additional support from experimental results taken by other means than mass spectrometry. The prompt mass yields, i.e., yields before neutron emission has

occurred, have been measured by simultaneous detection of the fission fragments with energy sensitive detectors. Figure 4 shows the prompt yield curve of the heavy fragments from ^{235}U fission (20). The mass spectrometric data on the cumulative yields are also shown for comparison. The prompt yield curve is quite smooth, indicating that the fine structure in the cumulative yield curve is the result of post-fission neutron emission. From the differences between these two curves, Terrell (21) has calculated the average number of neutrons emitted at each mass number. His neutron emission curve is also shown in Figure 4.

Figure 5 shows the neutron emission curve measured directly by Apalin et al. (22) for ^{235}U fission. It agrees well with Terrell's calculated curve. A close examination of these curves shows that the pronounced fine structure at mass 134 in the cumulative yield curve is generated by a modest change in the slope of the neutron emission curve between mass numbers 134 and 136. In conclusion, it now seems that fine structure in the cumulative yield curve results mainly from fluctuations in the neutron emission characteristics of the fragments, although closed-shell effects in the fission act itself may play a small part.

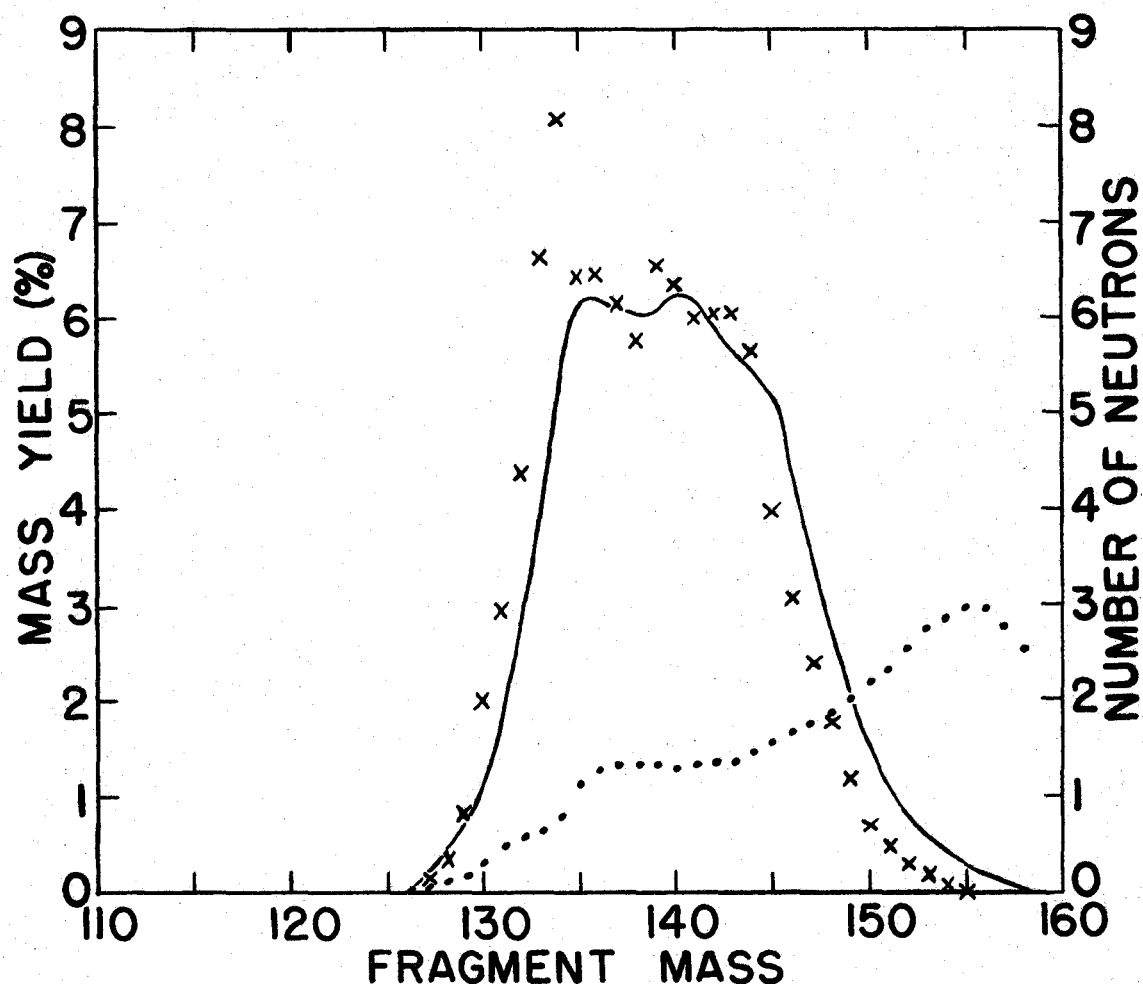


Fig. 4. Prompt mass yields and neutron emission characteristics as a function of the mass of the heavy fragment for the fission of ^{235}U . The solid line represents the prompt mass yield data of Fraser and Milton (20). The dots represent Terrell's calculations (21) for the number of prompt neutrons emitted at each mass number. For comparison, the cumulative yield data from Katcoff (23) are shown as crosses.

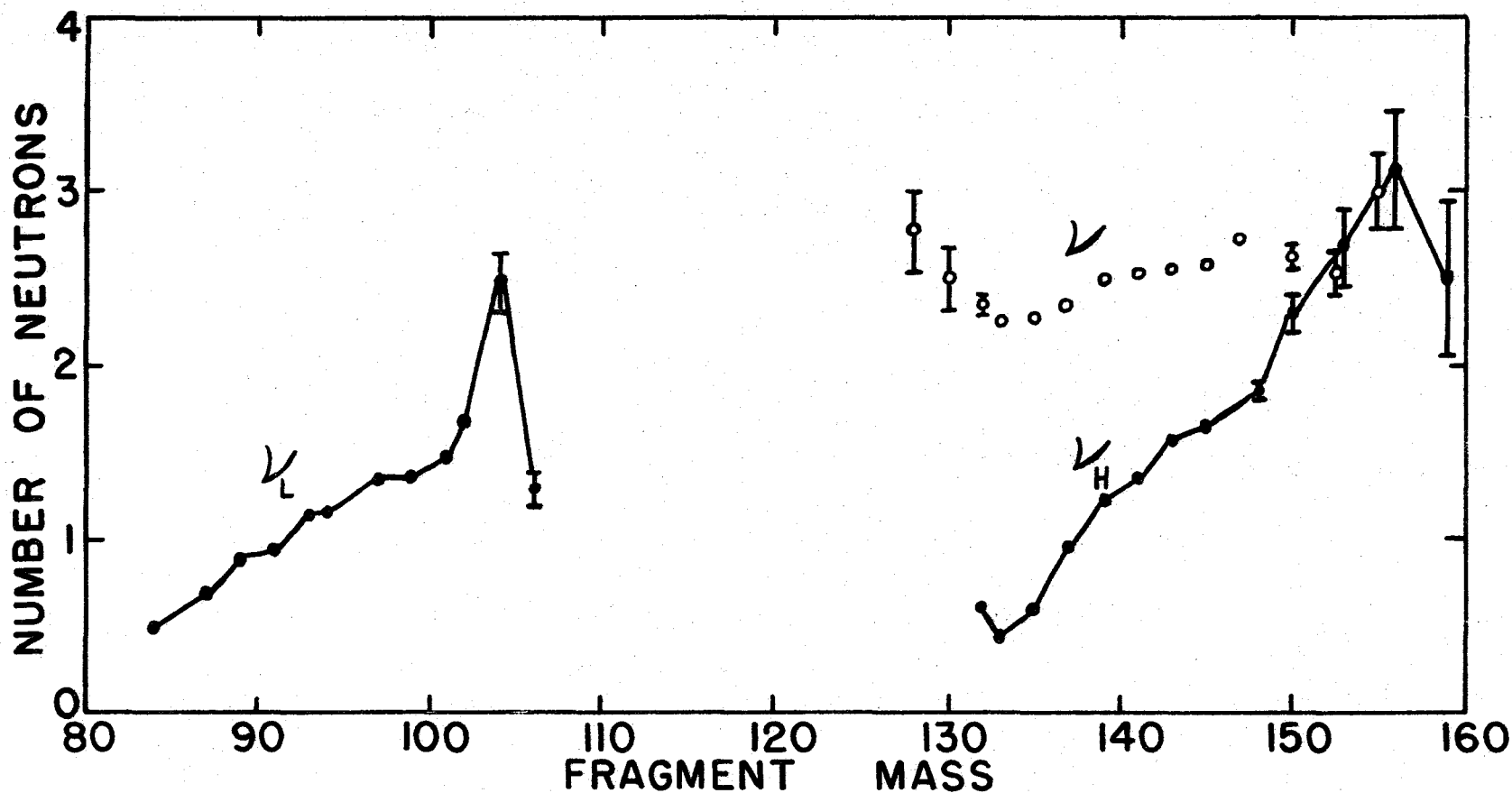


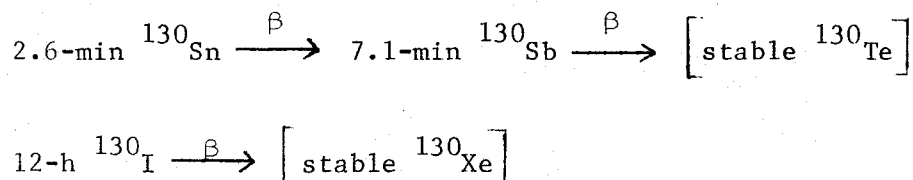
Fig. 5. Measurements by Apalin et al. (22) on the number of prompt neutrons emitted as a function of fragment mass for the fission of ^{235}U . The number of neutrons emitted from the light fragment, ν_L , and the number of neutrons emitted from the heavy fragment, ν_H , are shown as solid points. The open circles represent $\nu = \nu_L + \nu_H$, the total number of neutrons emitted as a function of the heavy fragment mass.

Irregularities in the cumulative yield curve can also result from chain-branching due to delayed neutron emission, or from neutron capture on certain fragments. A fission product may emit a delayed neutron if the β -decay of its precursor leaves it in a highly excited state. For example, 15 percent of the 4.4-second ^{89}Br formed in fission β -decays to an excited state of ^{89}Kr which immediately emits a neutron. This considerably alters the observed 88 and 89 yields. The fission fragment ^{135}Xe has an extremely large neutron capture cross section (2.7 million barns). Thus, if a sample is irradiated in an intense neutron flux, the observed ^{136}Xe yield is considerably enhanced. One usually makes corrections for these two effects, rather than regarding them as instances of true fine structure.

INDEPENDENT YIELDS

Most of the primary fission products have half-lives of a few minutes or less. Hence independent yield measurements must be carried out quickly, usually by radiochemical methods, and thus are often inaccurate. However, if a fission product nuclide happens to be shielded from β -decaying precursors by a stable nuclide, then its independent yield can be accurately measured by means of a mass spectrometer. An example occurs in the 130 mass chain, where ^{130}I

is shielded by stable ^{130}Te :



Here the total yield of ^{130}Xe is equal to the sum of the independent yields of ^{130}I and ^{130}Xe . However, in the region of stable nuclides, the independent yields fall off very rapidly with increasing atomic number and the independent yield of ^{130}Xe is less than one percent of the ^{130}I yield. Hence, if sufficient time is allowed for the decay of 12-hour ^{130}I , then the measured abundance of stable ^{130}Xe gives, quite accurately, the independent yield of the shielded nuclide ^{130}I .

For most fission product mass chains, the independent yields of only one or two members have been measured. The available data indicate that in the act of fission the nuclear charge is distributed along a given mass chain according to a Gaussian function of the following form:

$$P(Z) = \frac{1}{(\sigma\pi)^{\frac{1}{2}}} \exp \left[- \frac{(Z-Z_p)^2}{c} \right]$$

$P(Z)$ is the probability of forming a fragment with atomic number Z at a given mass number. This expression is normalized so that if the yields at all values of Z along the given mass chain are summed, the result is unity. The quantity c is related to the standard

deviation σ of the individual yields by the following equation:

$$c \approx 2 \left(\sigma^2 + \frac{1}{12} \right)$$

Usually, c is assumed to be constant for all mass chains, and is obtained from a best fit to several chains.

The most probable charge, Z_p , is usually fractional and varies with the mass number, A . Generally, some postulate is made, either on theoretical or else purely empirical grounds, in order to calculate Z_p as a function of A . If the constant c is known, then just one accurately measured independent yield on a given mass chain is sufficient to test a Z_p postulate. The most important Z_p postulates have been the following:

(1) Uniform Charge Distribution (UCD)

It might be expected that the most probable division of nuclear charge in fission would leave both fragments (before neutron evaporation) with the same charge density as the parent nucleus. In this case

$$Z_p = A \frac{Z_f}{A_f}$$

where Z_f and A_f refer to the parent nucleus. Actually, the experimental data indicate that the light fragment receives slightly more of the nuclear charge than the uniform charge distribution hypothesis would predict. This is not surprising, since stable light fragments have higher charge densities than stable heavy fragments. Nevertheless, the

uniform charge distribution postulate is useful in explaining the variation in Z_P at a given A for different fissioning nuclides.

(2) Equal Charge Displacement (ECD)

In 1949, Glendenin, Coryell and Edwards (24) were able to account for most of the independent yield data then available by assuming that, for a given mass split, the most probable charge, Z_P , is displaced from the most stable charge, Z_A , by equal amounts on both fragments. This can be expressed as

$$Z_P - Z_A = Z_P^* - Z_A^*$$

where the asterisk refers to the complementary fragment. Since nuclear charge must be conserved,

$$Z_P + Z_P^* = Z_f$$

$$\therefore Z_P = Z_A - \frac{1}{2}(Z_A + Z_A^* - Z_f)$$

Z_f is the atomic number of the fissioning nucleus. Z_A , the most stable charge at mass A , is usually fractional and can be calculated from a semi-empirical mass equation. Pappas (25) later extended this treatment by including shell effects in the calculation of Z_A , and obtained better agreement with experimental data. It should be noted that the mass number A and its complementary mass number refer to the fragments before prompt neutron emission has occurred. Hence, in order to make exact calculations of independent fission yields, one must have a knowledge of the number of prompt neutrons emitted

as a function of fragment mass.

(3) Maximum Total Energy Release

In 1956 Kennett and Thode (26) measured the independent yields of the shielded nuclides ^{128}I and ^{130}I from a number of fissioning species. They found that although the ^{130}I yields could be accounted for by equal charge displacement, the ^{128}I yields were too high by as much as a factor of 50. The two authors suggested that nuclear shell effects might influence the most probable charge, Z_p , as well as the most stable charge, Z_A . In order to calculate Z_p as a function of A , they postulated that the most probable mode of fission is that which gives the maximum release of total energy. Now the energy release in fission is equal to the so-called "mass deficit", i.e., the difference in mass between the fissioning nucleus and fission products. In symbolic terms,

$$E = M(Z_f, A_f) - \bar{\nu}M(n) - M(Z, A) - M(Z_f - Z, A_f - A)$$

M refers to the mass of the nuclear species contained in brackets and $\bar{\nu}$ is the average number of neutrons emitted in fission. The most probable charge, Z_p , is that value of Z which maximizes the expression for E . The masses of short-lived primary fission products cannot be measured directly. Kennett and Thode calculated these masses from a formula by Kumar and Preston (27) which includes shell effects. The resulting Z_p function or ^{235}U fission is shown in Fig. 6. The equal charge displacement (ECD) prediction for Z_p is shown as a

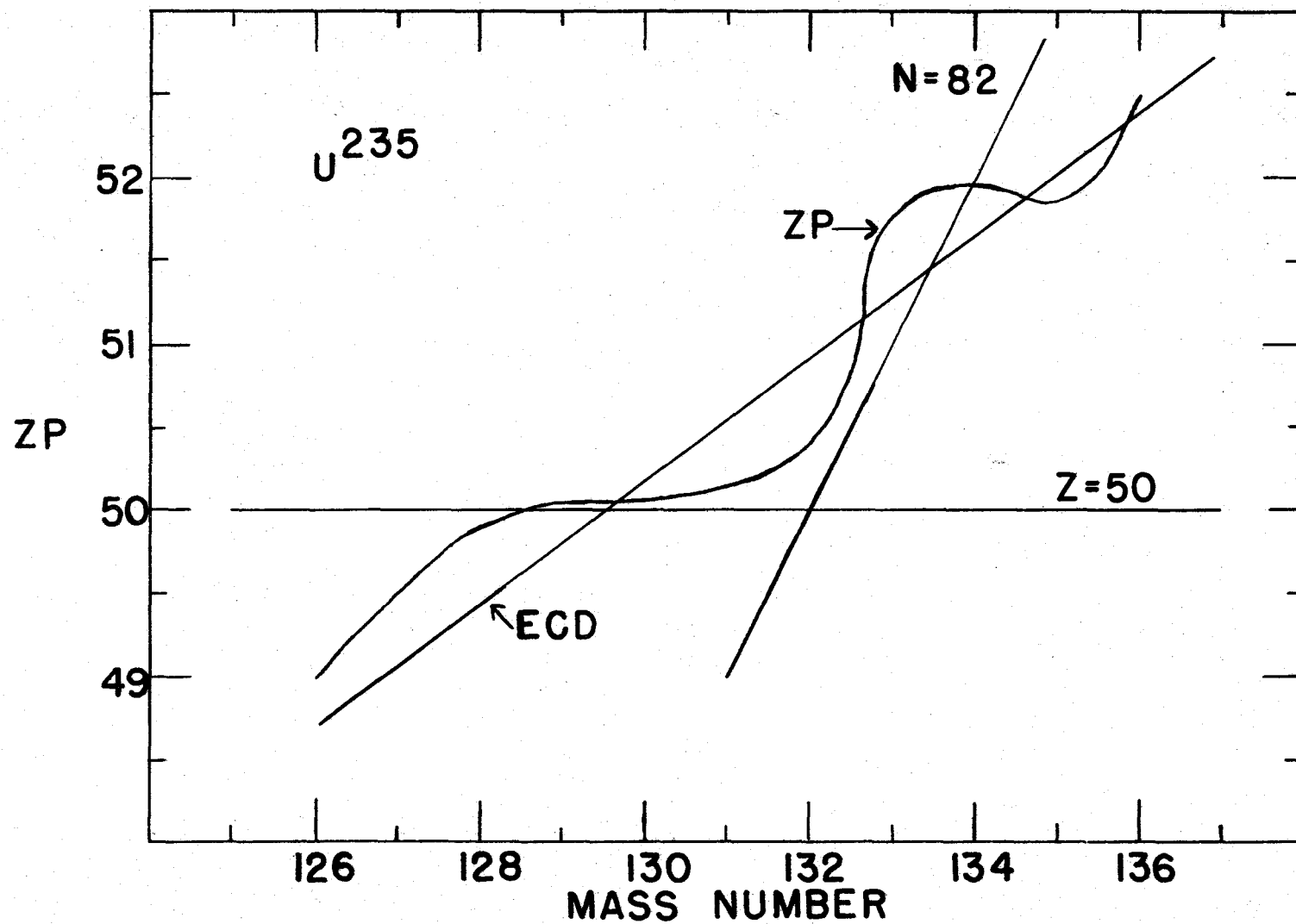


Fig. 6. The most probable charge, Z_p , calculated by the postulate of maximum energy release as a function of mass number. Also shown for comparison is the most probable charge calculated by the postulate of equal charge displacement (ECD).

solid line for comparison. Notice how the ECD line dips below the 50-proton shell at mass 128, while the maximum energy release curve remains above this shell. This upward shift in the value of Z_p is sufficient to account for the high yields of ^{128}I .

Alexander and Coryell (28) applied the maximum energy release postulate of Kennett and Thode to calculate Z_p for the entire mass range. They found that generally these Z_p values gave poorer agreement with experiment than did the original ECD predictions. Nevertheless, this approach does indicate the importance of shell effects in nuclear charge distribution.

(4) Maximum Excitation Energy

More recently, Wing and Fong (29) have applied the statistical theory of fission* to nuclear charge distribution. Utilizing their own nuclidic mass formula, they deduced the values of Z_p that maximize fragment excitation energies over the entire mass range of fission products for ^{235}U and ^{252}Cf fission. They then showed that their predicted Z_p values give a smaller root-mean-square deviation from experimental data than do most other schemes of charge distribution. It must be emphasized, however, that no theory of charge distribution gives complete agreement with all independent yield data.

In 1962 (30), and more more recently in 1966 (31), A. C. Wahl and co-workers, using refined radiochemical techniques, were able to

* See next section on Theories of Fission.

obtain two or three good independent yield measurements for each of ten different mass chains in ^{235}U fission. Their results confirmed that the distribution of nuclear charge in fission is best represented by the Gaussian function discussed previously. Within the limits of experimental error, they also found that distribution width, c , is indeed constant, as assumed in earlier treatments. These authors (30) applied their derived value of c to many other mass chains, each with only one known independent yield, and calculated the most probable charge, Z_p , for these mass numbers. They then derived an empirical $Z_p(A)$ function which is useful for testing theories of charge distribution and also for calculating independent yields in regions where experimental data are unavailable.

In 1966, P. O. Strom et al. (32) measured three independent yields at each of the mass numbers 131, 132, and 133. They fitted these data with a Gaussian distribution and found that c , rather than being constant in this region, decreased monotonically with mass number. The differences in their respective values of c were much too large to be attributed to experimental error. These results do not necessarily contradict Wahl's data, however, because the latter were taken in different regions of the mass yield curve.

Recently, E. Konecny et al. (33) have reported a method, involving β -counting, which gives the entire charge distribution curve at each mass number. These authors obtained distribution curves in the mass range 132-137 which are not gaussian but skewed. However, many refinements are needed in this technique before it can compete with the accuracy of older methods.

THEORIES OF FISSION

A complete theory of fission should explain the observed cumulative and independent yields described in the previous sections. It should also account for the kinetic energies, angular correlations, and excitation energies of all the fission products. Such a theory is far from being realized. The extreme complexity of fission precludes a complete solution of the dynamics of the fissioning nucleus. However, a number of simplified models have been proposed in order to explain the main features of the fission process. Some of the more important models are described here.

The Liquid Drop Model

Shortly after the discovery of fission, Bohr and Wheeler (34) suggested an analogy between the nucleus and a charged liquid drop. The "surface tension" which holds the drop together is provided by the strongly attractive nuclear forces. At the same time, the positive nuclear charge provides a repulsive coulomb force which tends to break up the nucleus into smaller droplets. The magnitude of the coulomb repulsion energy increases rapidly with nuclear size until it becomes equal to the surface tension energy. Nuclei with sizes greater than this critical size are unstable with respect to spontaneous disintegration. For heavy nuclei like ^{235}U , the surface tension energy is only slightly greater than the coulomb energy. All that is required to produce fission is a small excitation, which

can be provided merely by the binding energy gained in capturing a thermal neutron. This excitation energy manifests itself as oscillations in the nuclear shape. If the resulting distortion passes a certain critical point (the saddle point) the long range coulomb forces become dominant, causing the nucleus to split into two distinct fragments, which now repel one another and attain high kinetic energies.

Although this model gives us a qualitative understanding of fission and explains the relative stabilities of heavy nuclei, it fails to account for the predominantly asymmetric mass split in fission. To obtain a better explanation, one must consider the internal structures of the fissioning nucleus and resulting fragments.

Statistical Theory

The heavy mass peak of the cumulative yield curve occurs in the region of both the 82-neutron and the 50-proton shells. The further observation that this peak remains fixed as the fissioning nucleus is varied strongly suggests that nuclear shell structure is important in determining the division of nuclear mass. However, it is one thing to point out interesting correlations between magic numbers and mass distributions, and quite another thing to explain just how fragments, yet unformed, are able to influence the direction of the fission process.

In 1956 Fong (35) provided such an explanation with his statistical theory of fission. He postulated that the fission act occurs slowly enough to maintain thermodynamic equilibrium throughout the fissioning nucleus right up until the moment of scission. Thus the probability of obtaining a given set of fission fragments is proportional to the densities of internal excitation states of those particular fragments at the moment of scission. The density of states, $W_0(E)$, is related to the internal excitation energy of the fragments, E , by the following expression:

$$W_0(E) = c \exp \left[2 (aE)^{1/2} \right]$$

Fong obtains this excitation energy by subtracting the coulomb energy and deformation energy of the fragments from the total energy release. The parameters "c" and "a" are obtained from fits with neutron absorption data.

Nuclei in the region of closed shells have lower ground state energies than those far removed from closed shells. Thus such closed-shell nuclei, when they are formed in fission, will have slightly higher excitation energies. Now a small difference in excitation energy leads to a large difference in the density of states, and hence in the probability of formation. Thus, qualitatively at least, we see how the shell structure of the fragments can influence the fission process in favour of asymmetric splitting. More excitation

energy is available for the asymmetric mode.

Fong carried out calculations for ^{235}U fission and was able to reproduce the general asymmetric shape of the observed mass distribution. However, attempts to apply his theory to other fissioning nuclides have not met with similar success (5). Also, it is now believed that the fissioning nucleus is very "cold" at the saddle point, i.e., only a few excitation states are available at this point. If this is so, then Fong's statistical expression for the density of states is no longer valid.

Asymmetric Transition State

A. Bohr (36) has proposed that the main features of fission are determined by the quantum states of the nucleus at its saddle point rather than at the moment of scission. He argues that at the saddle point most of the excitation energy is present as deformation of the nuclear shape. Thus only a few internal excitation states are available to the compound fissioning nucleus, and these should resemble the low-lying states of the undeformed nucleus. Now even-even heavy nuclei show a 1^- rotational band near the ground state. This negative parity band has been associated with a stable octupole deformation (see Johansson (37)) which is an asymmetric shape. If the fissioning nucleus were to pass through such a negative parity transition state, then the asymmetry of the mass split would be determined at this point. Angular correlation

measurements between the bombarding particle which induces fission and the recoiling fission fragments show that the compound nucleus passes through only a small number of transition states. Unfortunately, no angular correlation data are available on fission induced by thermal neutrons.

This theory explains why symmetric fission becomes more probable as the energy of the bombarding particle is increased. At the higher excitation energy, more transition states become available to the compound nucleus, and the fission process is less dependent on the lower energy asymmetric states. Closed shell effects in the fragments may still play a part in the later stages of fission, as the compound nucleus descends from saddle point to scission.

Whetstone Model

Whetstone (38) has devised a model to account for asymmetric fission as well as for the saw-tooth variation of neutron emission as a function of fragment mass. (See Figures 4 and 5 on pages 12 and 13.) He represents the fissioning nucleus at its saddle point by an asymmetric dumb-bell shape, as shown in Figure 7. The two ends of the dumb-bell are nearly spherical, possibly influenced by shell structure. The neck is thin and rich in neutrons. The fragment, which after scission receives the major portion of the neck, will have a larger deformation energy and will emit more neutrons. In Figure 7 a mass split at position 1 corresponds to improbable symmetric fission. In this case most of the neutrons come from the

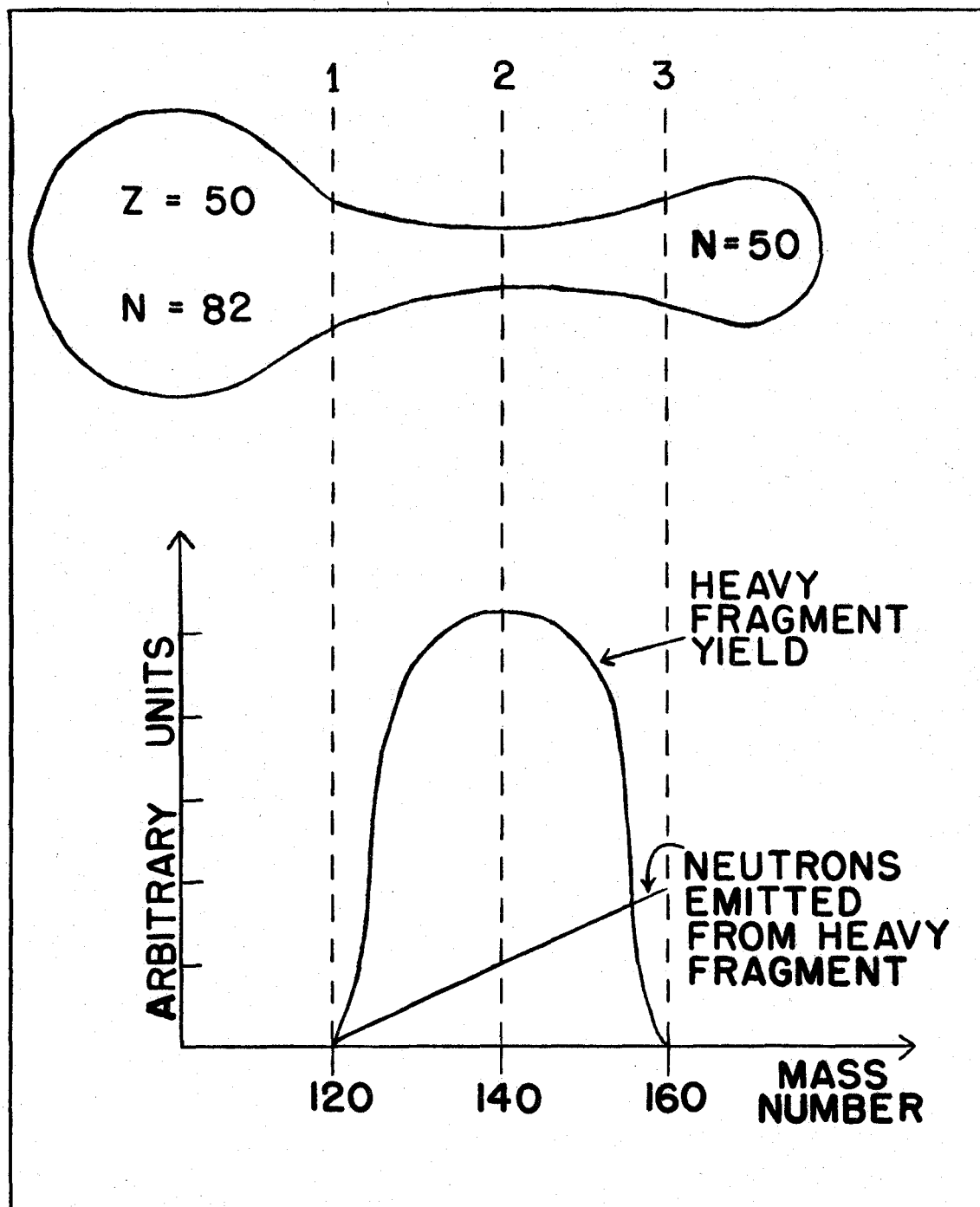


Fig. 7. The Whetstone model of the fissioning nucleus.

light fragment. The mass split at position 2, the thinnest point in the neck, corresponds to the most probable mode of fission. For the highly asymmetric split at position 3, most of the neutrons are emitted from the heavy fragment.

Although none of the above models gives a complete explanation of all fission yield data, each offers some insight into the fission process.

RARE GAS FISSION PRODUCTS

Rare gases do not tend to form chemical bonds. For this reason the rare gas fission products can easily be extracted from an irradiated sample and introduced in pure form into a mass spectrometer. The 83, 84, and 86 isotopes of krypton and the 131, 132, 134, and 136 isotopes of xenon represent the end products of seven mass chains produced in fission. In addition, the unstable 85, 87, and 88 isotopes of krypton and the unstable 133 and 135 isotopes of xenon give us, after appropriate corrections for partial decay, the cumulative yields of five more mass chains. These twelve mass chains represent over forty per cent of all fission products. The xenon yields lie on top of the heavy mass peak, where fine structure effects are most prominent. The krypton yields are on the low side of the light mass peak, and are very sensitive to small changes in

the position of this peak from one fissile nuclide to another. Thus, much valuable information about the mass yield curve can be obtained by studying the fission yields of the rare gases.

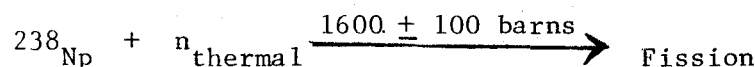
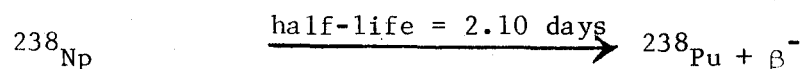
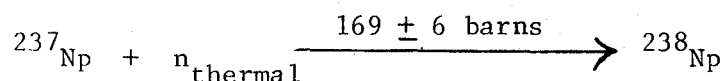
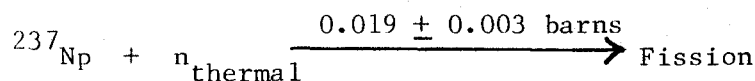
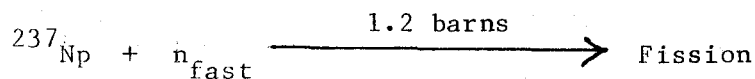
A mass spectrometric analysis of rare gas fission products can also provide accurate data on independent yields. The abundances of ^{80}Kr , ^{82}Kr , ^{128}Xe , and ^{130}Xe represent the independent yields of the shielded nuclides ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I . Accurate measurements of such independent yields are necessary in order to test theories of nuclear charge distribution in fission.

Cumulative Yields from the Fission of ^{237}Np and ^{238}Np

Mass spectrometric determinations of the cumulative xenon and krypton yields have been carried out for the thermal neutron fission of ^{235}U (6), ^{233}U (10), ^{239}Pu (7), ^{241}Pu (8), ^{241}Am (9), and ^{242}Am (9), and also for the fast neutron fission of ^{238}U (11) and ^{232}Th (13). No work has been done as yet on the rare gas yields from the fission of neptunium, although this transuranium element was discovered in 1944.

The longest-lived isotope of neptunium, ^{237}Np , has a half-life of 2.14 million years but its fission cross section for thermal neutrons is only 0.019 barns. However, when irradiated in a nuclear reactor, ^{237}Np captures neutrons to form ^{238}Np (half-life = 2.10 days) which does have a large fission cross section for thermal neutrons (1600 barns). The isotope ^{237}Np also undergoes fission with fast

neutrons, i.e., neutrons with energies in the Mev range. The neutron reactions of interest in neptunium are as follows:



The only data available on neptunium fission yields have been obtained by radiochemical methods. The first such investigations were carried out in 1957 by Ford and Gilmore (39), who measured the yields from the fission of ^{237}Np by fission spectrum neutrons (average energy = 1.1 Mev). Coleman, Hawker, and Perkin (40) later studied the same reaction with 14.5 Mev neutrons. The mass yield curves from both investigations are quite similar to those resulting from the fission of ^{235}U , ^{238}U , and ^{239}Pu by neutrons in the same energy range. More recently, the fission of ^{237}Np by fission spectrum neutrons has been studied by I. S. Iyer et. al. (41) and

by R. Stella et al. (42). Their mass yields confirm the general trend that, as the mass of the fissioning nucleus increases, the heavy mass peak remains fixed while the light mass peak shifts to higher mass numbers. However, the precision of these radiochemical measurements is not great enough to reveal fine structure in the mass yield curve.

No studies have been made of the yields from the fission of ^{237}Np by neutrons of thermal energy, and no data at all are available on the yields from ^{238}Np fission. Accurate mass spectrometric analyses of the products from the fission of ^{237}Np and ^{238}Np would increase our knowledge of how the mass yield curve differs from one fissile nuclide to another. This thesis describes such analyses. The cumulative xenon yields in the range of masses 131 to 136 and the cumulative krypton yields in the range of masses 83 to 88 have been measured for the thermal neutron fission of both ^{237}Np and ^{238}Np , as well as for the fission of ^{237}Np by fission spectrum neutrons. Only the relative yields of xenon and krypton isotopes have been measured. Uncertainties in the fission cross sections and lack of information on yields in other regions of the mass yield curves have precluded accurate absolute yield determinations.

The results are compared with those from other fissioning nuclides in order to clarify general trends in the mass yield curve.

Independent Yields of ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I from Various Fission Processes

The first measurement of the independent yield of a shielded nuclide was reported by Feldman, Glendenin, and Edwards in 1945 (43). This was a radiochemical determination of the ^{82}Br yield from the thermal neutron fission of ^{235}U . A decade later Kennett and Thode (26, 44) were able to measure, by means of mass spectrometric analyses of rare gas fission products, the independent yields of the four shielded nuclides ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I from the fission of ^{235}U , ^{233}U , and ^{239}Pu . They obtained ^{130}I yields that were in reasonable agreement with predictions based on the postulate of equal charge displacement. However, they found the ^{128}I yields to be higher than the predictions by as much as a factor of 100. Since ^{128}I can also be formed by neutron capture on any natural iodine that might be present in the fissionable material, it would be desirable to confirm these anomalously high yields by independent measurements. In addition, serious doubts have been cast on the yields of ^{80}Br and ^{82}Br obtained by Kennett and Thode, because of the possibility of contamination by natural bromine or krypton.

With the above considerations in mind, the independent yields of the four shielded nuclides ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I have been redetermined in the present work for the thermal neutron fission of ^{235}U , ^{233}U , and ^{239}Pu . Data have also been obtained for the ^{82}Br ,

^{128}I , and ^{130}I yields from ^{238}Np fission, and an upper limit has been set for the independent yield of ^{126}I from ^{235}U fission. These yields have been obtained by mass spectrometric analyses of the respective xenon and krypton daughters of the shielded nuclides. Great care has been taken to eliminate natural contamination from the samples.

The independent yield results are examined in terms of conventional charge distribution theories and also in terms of neutron emission effects from prompt fission fragments.

EXPERIMENTAL PROCEDURE

SAMPLE PREPARATION

Uranium

Both ^{235}U and ^{233}U were used in the study of independent bromine and iodine yields. The ^{235}U was obtained in the form of U_3O_8 which was enriched to 93.18% in the 235 isotope. This compound was readily dissolved in warm, concentrated nitric acid. The ^{233}U had been used in a previous experiment and was recovered from an ion exchange resin. Alpha counting showed this uranium to be essentially pure ^{233}U .

It was necessary to reduce the natural bromine and iodine contamination in the uranium samples to a minimum. Neutron capture on the stable bromine and iodine isotopes leads to the formation of ^{80}Br , ^{82}Br , and ^{128}I --nuclides whose independent yields were to be measured. For example, the neutron capture on one part natural bromine per million parts ^{235}U would produce as much ^{80}Br as is produced directly in fission. Twenty parts iodine per million parts ^{235}U would lead to an amount of ^{128}I equal to that produced directly in fission. Thus purification procedures were developed for eliminating traces of natural bromine and iodine from uranium.

Most of the samples were purified by hydrogen peroxide

which precipitated the uranium from solution while leaving any bromide or iodide behind in solution. The uranium was first dissolved in nitric acid, and this solution was evaporated to dryness. The residue was dissolved in water, evaporated to dryness, and redissolved, etc., until a neutral solution of uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2$) was obtained. Then the uranium was precipitated with H_2O_2 and the precipitate was separated from solution and washed with water. The precipitate was then dissolved in HNO_3 and the cycle was repeated.

The effectiveness of this procedure was tested by adding 36-hour ^{82}Br to a nitrate solution of natural uranium. The radioactive bromine was produced by irradiating several milligrams of NH_4Br in the McMaster Nuclear Reactor for about 15 minutes. After two hydrogen peroxide cycles, gamma counting measurements showed that the level of bromide contamination in the natural uranium had been reduced from 1 part per 100 to less than 300 parts per billion, i.e., a decontamination factor of greater than 30,000 had been obtained. No measurements of iodide decontamination were carried out for the hydrogen peroxide procedure, although it was assumed that any iodide would be removed from the uranium along with the bromide.

Samples of purified uranium were evaporated onto pieces of aluminum foil, each about one centimeter square. The purpose of the foils was to absorb and hold recoiling fission fragments. The

dry uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) had to be heated in order to drive off the water of crystallization, which would have caused "sputtering" of the samples during subsequent heating. Each foil held about one milligram of uranium and was carefully rolled up and dropped into an irradiation ampoule, such as the one shown in Figure 8. These ampoules were made of quartz since ordinary glass deteriorates under irradiation. The ampoules were then evacuated on a high vacuum line and the foils outgassed by being heated to a dull red colour. This was necessary in order to remove any traces of atmospheric krypton and xenon from the samples. Each ampoule was sealed off, removed from the vacuum line, and leak-tested by observing the Tesla coil discharge inside it. Finally, for each irradiation, an ampoule was placed inside an aluminum can (length - $1\frac{3}{4}$ "; diameter - $\frac{7}{8}$ ") whose lid was cold-welded into place.

Four samples of ^{235}U and four of ^{233}U were irradiated in the McMaster Nuclear Reactor for periods varying from two to six weeks, so as to produce between 10^{16} and 10^{17} fissions in each sample. It was desirable to obtain the largest possible number of fissions in order to measure precisely the very low independent bromine and iodine yields. However, this led to highly radioactive samples, which could not be handled safely until several months had elapsed. By this time the mass chains of interest had almost completely decayed to stable isotopes of xenon and krypton.

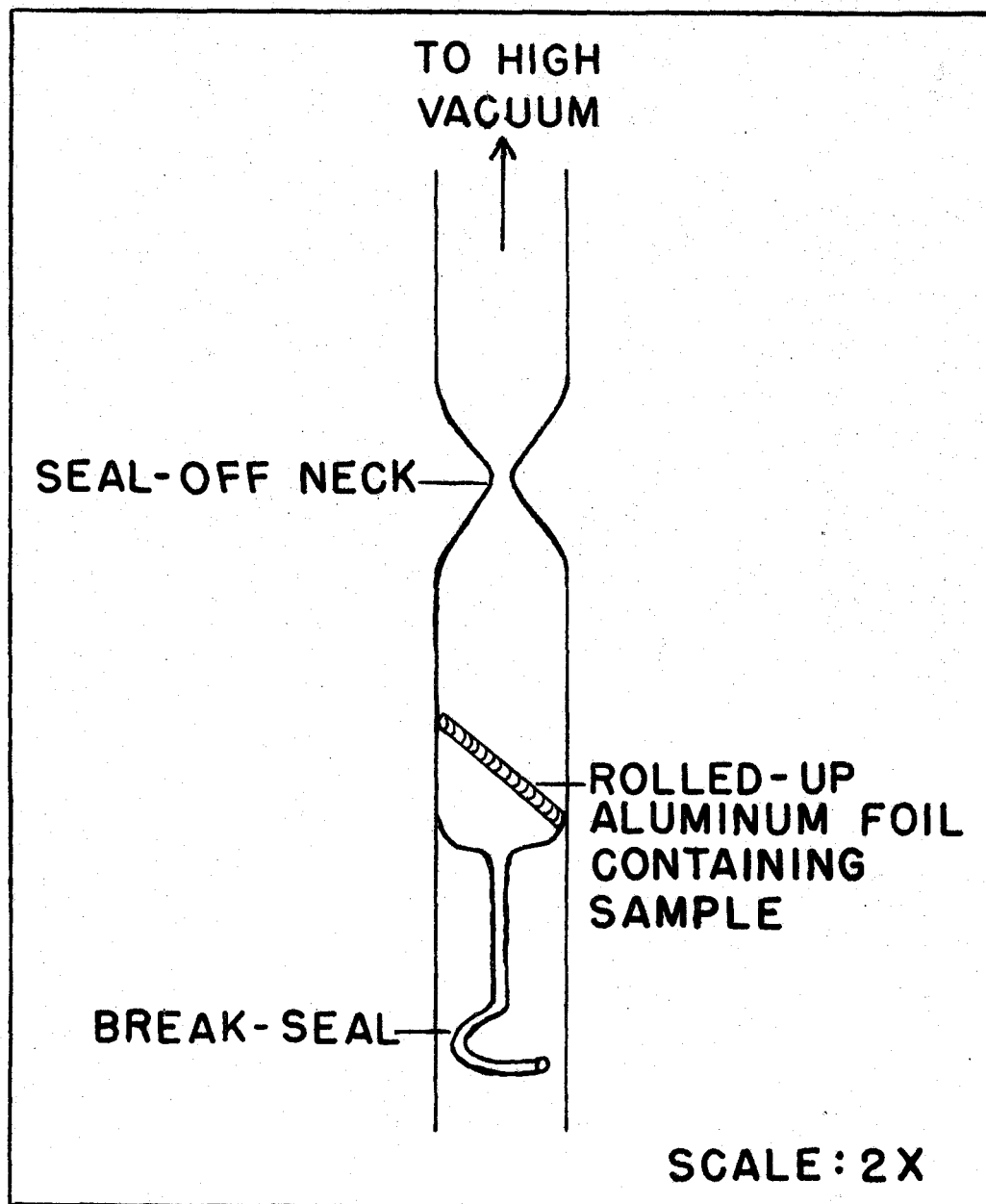


Fig. 8. Ampoule containing uranium sample.

The procedure described below for extracting the rare gases was the same for most of the fissionable nuclides studied. The quartz ampoule containing the irradiated sample was attached to a high vacuum line in the radioisotope laboratory. Figure 9 shows the general arrangement. After the line was thoroughly evacuated, the breakseal on the ampoule was broken by means of an iron slug manipulated with magnets. The fission product gases were extracted by heating the foils to a bright orange colour for about ten minutes. The heating was performed with an oxygen-natural gas flame. This extraction procedure was sufficient to remove at least 90% of the rare gases. The evolved gases were purified in a titanium furnace and then condensed in a gas sample tube on a charcoal trap cooled by liquid nitrogen. Finally the gas sample tube was sealed off, removed from the high vacuum line, and attached to the mass spectrometer line.

In spite of all precautions, many of the irradiated samples were found to contain traces of atmospheric xenon and krypton. Since this contamination did not occur in irradiated blanks, it was concluded that the quartz ampoules containing fissile nuclides had been damaged during irradiation -- either by recoiling fission products or by excessive heating from the fission reaction. Thus a small quantity of air had been able to enter the ampoules. However, it proved possible by a modification of the extraction procedure to separate most of the atmospheric gases from the fission

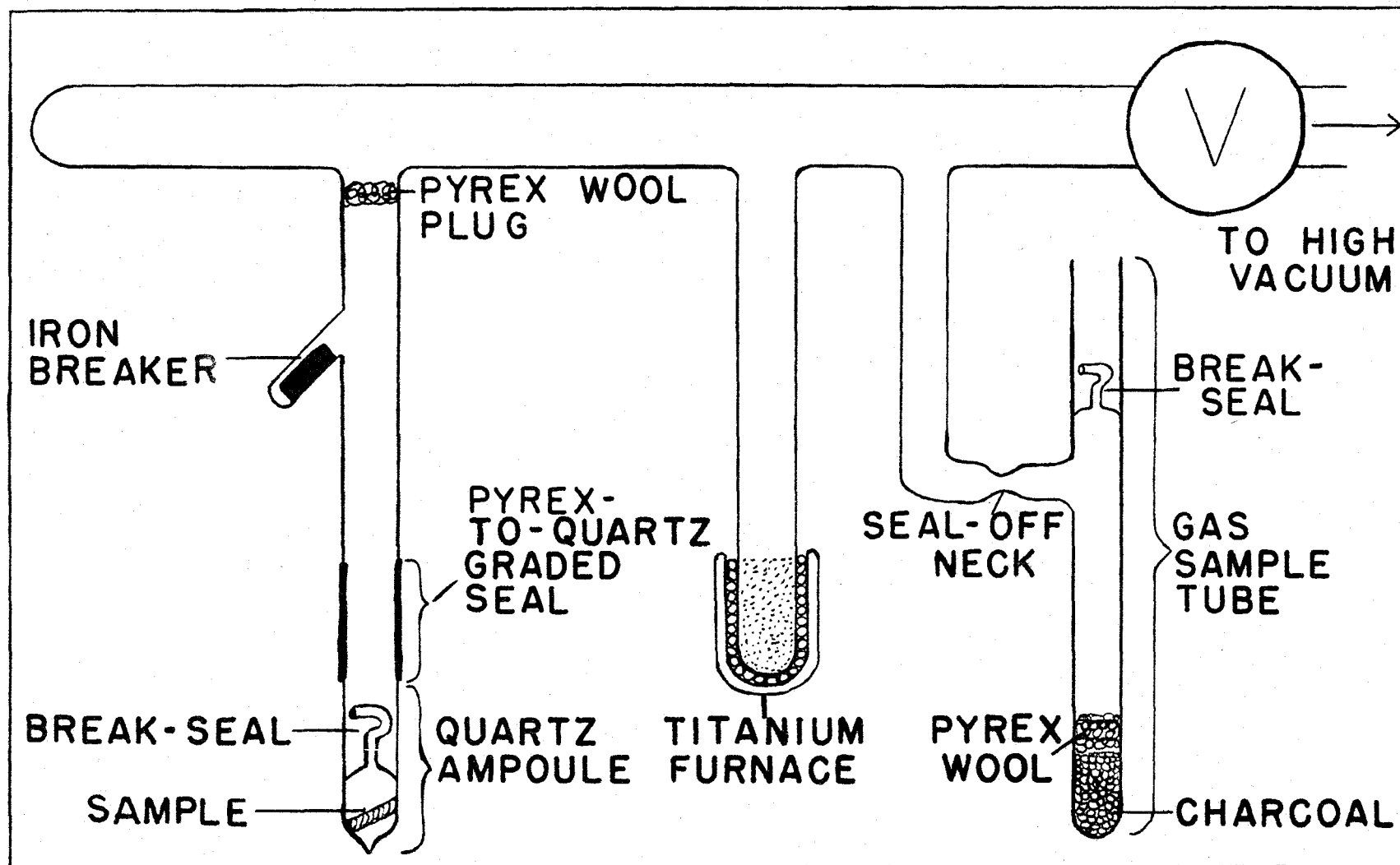


Fig. 9. Apparatus used for the extraction and purification of the rare gases.

product gases. Over 90% of the latter remained embedded in the aluminum foil or alloy, while the atmospheric gases were free inside the ampoule. After the break-seal to the ampoule was broken, the sample was gently heated and pumped on for several minutes. Then the valve to the pump was closed and the fission products were extracted. In this way the major portion of the fission product rare gases were recovered with little or no air contamination.

Four ^{235}U fission samples were prepared by a different method. $^{235}\text{U}_3\text{O}_8$ powder was placed in quartz irradiation ampoules which were then attached to a high vacuum line. While being pumped out, the ampoules were heated in an induction furnace to a temperature of 1500°C for three hours. This heating expelled all volatile impurities, but left behind the highly refractory U_3O_8 . The ampoules were then sealed off and irradiated in the McMaster Reactor for several months. This produced up to 10^{18} fissions in each sample. After a convenient period of time, the rare gases were extracted from the samples, again by heating in the induction furnace. The chlorine contamination in these samples was estimated to be less than 20 parts per billion. One would expect any contamination by the less abundant bromine and iodine to be at an even lower level.

Plutonium

The ^{239}Pu for the independent yield studies was obtained in the form of an alloy (14% plutonium, 86% aluminum). Because of the

hazardous α -activity of ^{239}Pu (half-life = 20,000 years) it was considered desirable to minimize handling of the samples. Consequently, no chemical purification procedures were carried out on the plutonium. However, the plutonium alloy has a much lower melting point than that of the uranyl nitrate discussed previously. Heating the alloy to its melting point should be sufficient to expel any bromine or iodine contamination. The effectiveness of this procedure was later verified by the consistency of independent yield results from one plutonium sample to the next.

Flakes of the alloy were weighed out and placed inside a small quartz tube which was stoppered with a quartz wool plug. The tube was then inserted into a quartz irradiation ampoule, as shown in Figure 10. It was found that if the alloy were placed directly in contact with the outer ampoule wall, the latter tended to crack upon subsequent heating. Thus the inner quartz tube was necessary. As the ampoule was being pumped out, the alloy was heated to a dull red colour in order to expel impurities. The ampoule was then sealed off, leak-tested, and placed in an aluminum irradiation can.

Five ^{239}Pu samples were irradiated in the McMaster Nuclear Reactor for periods varying from ten to twenty-eight days. This was sufficient to produce between 10^{16} and 10^{17} fissions in each sample. The rare gas fission products were subsequently extracted in a manner similar to that described for the uranium samples.

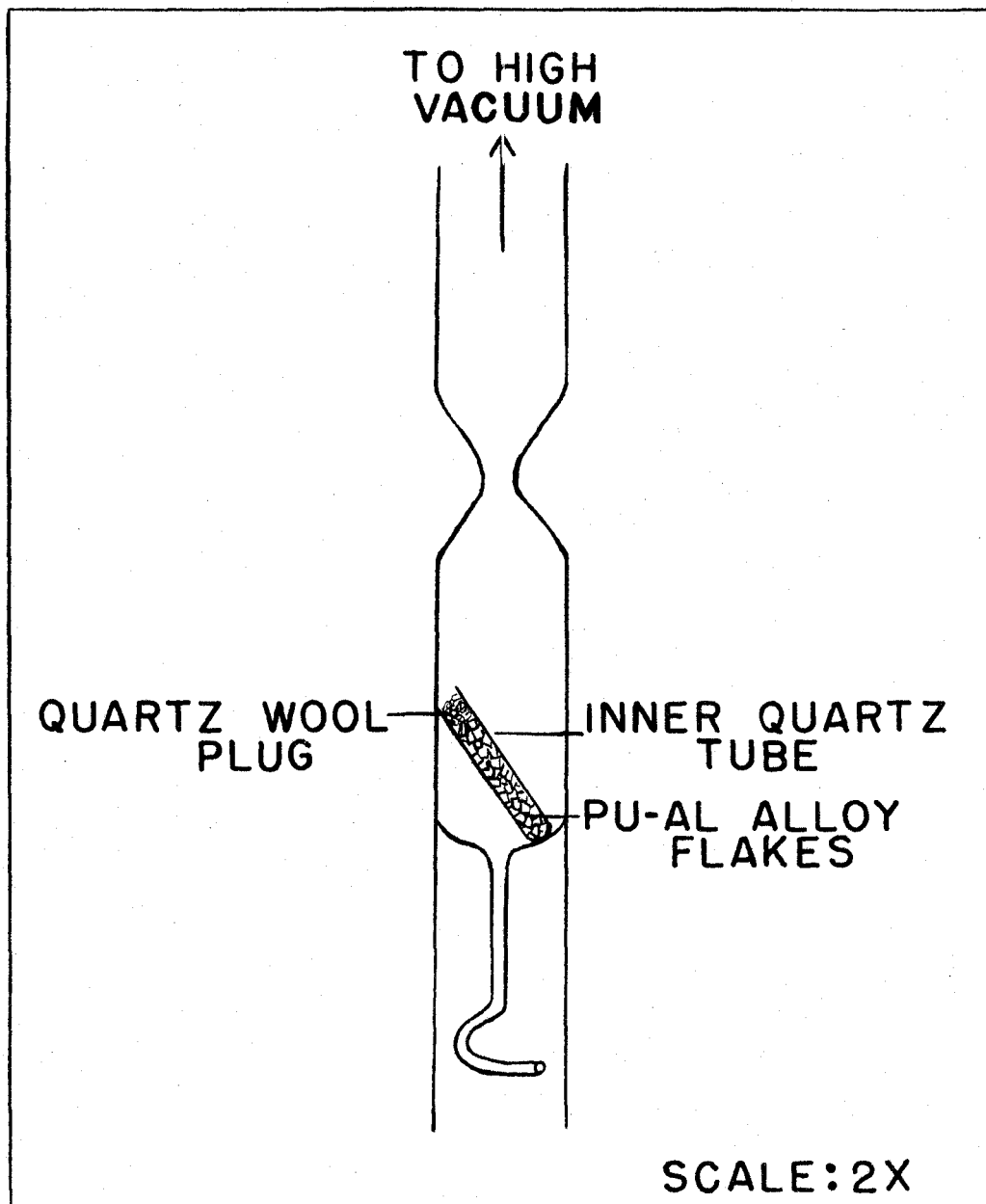
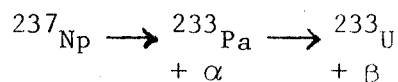


Fig.10. Ampoule containing plutonium sample.

Neptunium

A sample of 100 mg of $^{237}\text{NpO}_2$ was obtained from Oak Ridge National Laboratories. Concentrated HNO_3 and HCl were found to be ineffective in dissolving the NpO_2 . It was readily dissolved, however, in pure water after having been fumed for several hours with concentrated H_2SO_4 .

It is important to analyze the neptunium for traces of other fissile nuclides which might contribute to the fission product spectrum. For example, if ^{239}Pu were present in the neptunium to the extent of 0.1%, then in a neutron flux of 1×10^{13} neutrons/cm²/sec the ^{239}Pu fission rate would equal that of the ^{238}Np and would be 35 times as great as that of the ^{237}Np . The presence of ^{235}U and ^{233}U would be equally serious. There are reasons to suspect that these highly fissile nuclides might be present in the neptunium. ^{237}Np is produced from the neutron irradiation of uranium enriched in the ^{235}U isotope. ^{239}Pu is also a product of this irradiation. Thus an incomplete chemical separation of the neptunium might leave traces of ^{235}U or ^{239}Pu in the sample. Also, ^{237}Np decays to ^{233}U by the following sequence:



If the neptunium sample had been allowed to stand for several years, enough ^{233}U would have accumulated to seriously affect the ^{237}Np fission results.

The α spectrum of the neptunium sample was analyzed for possible contamination. The analysis revealed the presence of 89-year ^{238}Pu to the extent of 0.1%. This probably resulted from the β -decay of 2.10-day ^{238}Np , which had been present in the neptunium sample when it was purified at Oak Ridge. The thermal neutron fission cross section of ^{238}Pu is about 1,000 times as great as that of ^{237}Np . Hence an irradiation of the unpurified neptunium would produce as many ^{238}Pu fissions as ^{237}Np fissions.

With regard to other possible contaminants, the alpha group of 2×10^4 -year ^{239}Pu was not observed, and the upper limit of ^{239}Pu contamination was set at 1%. This is still high enough to be serious. The ^{235}U activity is too low to be observed in such an analysis, and no conclusions could be drawn about its presence. The alpha group from ^{233}U coincides with that from ^{237}Np . However, a separation of neptunium and uranium was carried out, and an analysis of the uranium fraction showed the ^{233}U contamination to be less than 0.003%, which is an acceptable level.

Because of the observed ^{238}Pu contamination and also because of the possibility of serious ^{239}Pu or ^{235}U contamination, it was necessary to purify the neptunium sample. An ion exchange procedure, described by Nelson, Michelson, and Holloway (45) for the separation of uranium, neptunium, and plutonium, was adopted with some modifications. A quantity of the dissolved neptunium was taken up in a solution of 9M HCl -- 0.05M HNO_3 and added to an anion exchange column (Dowex 1-X10

Resin, 200-400 mesh). The neptunium was adsorbed in the (IV) oxidation state along with any plutonium as Pu(IV) and uranium as U(VI). Any thorium, americium, or other fissile actinides would have passed through the column unadsorbed at this stage. Then the plutonium was eluted with 9M HCl -- 0.05M NH_4I . The iodide was added to reduce the plutonium to the non-adsorbable (III) state while leaving the neptunium and uranium unaffected. The column was thoroughly washed with 9M HCl and the neptunium eluted with 4M HCl -- 0.1M HF. The resin, now containing only uranium, was discarded. The purity of the neptunium fraction was tested by α -counting and repurification was carried out if required. By this procedure the ^{238}Pu contamination was reduced from 0.1% to less than 0.0004%. Any ^{239}Pu contamination should have been reduced by an equal factor.

If the neptunium sample was intended for independent bromine and iodine yield studies, then an additional step was necessary in order to remove any residual iodide used in the reduction of the plutonium. The sample was re-adsorbed on a fresh column and thoroughly washed with 9M HCl. Then the neptunium was eluted with 4M HCl -- 0.1M HF as before. Gamma counting measurements using 8-day ^{131}I as a tracer showed that this step reduces the iodide contamination to less than five parts per billion parts neptunium.

After purification the neptunium was converted to nitrate form and evaporated onto aluminum foils. The subsequent handling of the neptunium samples and extraction of the rare gases was similar

to the procedure already described for uranium. However, most of the irradiated neptunium samples had much lower activities than the irradiated uranium and plutonium samples. As a consequence, the extraction and purification of the rare gases from these neptunium samples could be carried out directly on the mass spectrometer line rather than in the radioisotope laboratory. The elimination of this extra step reduced the possibility of sample loss or contamination.

Special comments are required in discussing the neptunium irradiations. These irradiations were carried out under a variety of conditions in order to observe the fission products from three essentially different reactions:

1. The thermal neutron fission of ^{238}Np
2. The fast neutron fission of ^{237}Np
3. The thermal neutron fission of ^{237}Np

In order to produce thermal neutron fission of ^{238}Np , the sample must be irradiated in a high neutron flux for sufficient time to allow the growth of 2.10-day ^{238}Np by neutron capture, i.e., for at least several days. However, the irradiation time must not exceed one month, or else the fission of ^{238}Pu and ^{239}Pu would become serious. These nuclides are produced slowly by multiple neutron capture on ^{237}Np in a high neutron flux. Three samples were thus irradiated in the core of the NRU Reactor at Chalk River Nuclear Laboratories (flux = 2×10^{14} neutrons/cm²/sec) for a period of two weeks. In addition, five samples were irradiated in the McMaster Nuclear Reactor

(flux = 1.5×10^{13} neutrons/cm²/sec) -- one for 30 days, one for five days, and three for 60 hours each. These last three samples were analyzed within a few hours of irradiation in order to obtain the yields of several unstable isotopes -- 5.27-day ¹³³Xe, 9.2-hour ¹³⁵Xe, 76-minute ⁸⁷Kr, and 2.8-hour ⁸⁸Kr.

In order to produce fast neutron fission of ²³⁷Np, two samples were wrapped in cadmium and irradiated in the McMaster Nuclear Reactor for a period of four hours. Cadmium is almost completely opaque to thermal neutrons but is relatively transparent to fission spectrum neutrons. The use of cadmium thus inhibits the thermal neutron fission of ²³⁸Np, while allowing the fast fission of ²³⁷Np. Also, the irradiation time is short enough to prevent significant growth of ²³⁸Np in the sample.

It is difficult to observe the fission product spectrum from the thermal neutron fission of ²³⁷Np, since this nuclide has a thermal fission cross section of only 0.019 barns. At ordinary reactor fluxes, a long irradiation leads to the fission of ²³⁸Np, whereas a short irradiation produces fast fission of ²³⁷Np. What is required is a very low, well thermalized, neutron flux. This can be seen from the following expression, derived in Appendix A, for the ratio of ²³⁸Np fissions to ²³⁷fissions:

$$\frac{N_{238}^f}{N_{237}^f} = \frac{\sigma_f^{238} \sigma_c^{237}}{\sigma_f^{237} \lambda'} \phi \left(1 - \frac{1 - e^{-\lambda' T}}{\lambda' T} \right) \dots \dots \dots (1)$$

where

N_{238}^f = number of ^{238}Np fissions,

N_{237}^f = number of ^{237}Np fissions,

σ_f^{237} = ^{237}Np fission cross section for thermal neutrons = 19 mb,

σ_f^{238} = ^{238}Np fission cross section for thermal neutrons = 1600 ± 100 b,

σ_c^{237} = ^{237}Np capture cross section for thermal neutrons = 169 barns,

T = irradiation time,

ϕ = thermal neutron flux,

$\lambda' = \lambda + \sigma_f^{238} \phi$

λ = decay constant of 2.10-day ^{238}Np

At low fluxes, $\lambda' \approx \lambda$. After several days, the amount of 2.10-day ^{238}Np in the sample reaches equilibrium and the time-dependent term in equation (1) approaches unity. The ratio of ^{238}Np fissions to ^{237}Np fissions then becomes directly proportional to the thermal neutron flux. In a flux of 10^{13} neutrons/cm²/sec this ratio is about 37, but in a flux of 10^{10} neutrons/cm²/sec it is only 0.037.

The thermal column of the NRU Reactor at Chalk River has a well thermalized flux of 10^{10} neutrons/cm²/sec. Three samples were irradiated in this column, one for 70 days and two for 30 days, in order to produce thermal neutron fission of ^{237}Np . Because of the low flux and low fission cross section, the ^{237}Np fission rate is small. Thus the long irradiation times are required to build up

sufficient fission products for analysis.

Cobalt Flux Monitors

A knowledge of the neutron flux is necessary in the ^{238}Np fission irradiations for calculating the neutron capture corrections on ^{135}Xe , and in the ^{237}Np thermal fission irradiations for calculating the ratio of ^{238}Np fissions to ^{237}Np fissions from equation (1). Also, independent yield measurements of ^{82}Br , ^{128}I , and ^{130}I must be corrected for neutron capture contributions from the preceding mass chains. Thus cobalt flux monitors were irradiated with most of the samples. Each monitor consisted of a piece of 1% cobalt-aluminum wire. The resulting ^{60}Co activities were measured in an argon-filled ionization chamber whose output was amplified by a vibrating reed electrometer. The ionization chamber was calibrated by three standard ^{60}Co samples whose absolute disintegration rates had been previously determined by absolute beta counting. The neutron flux ϕ is given by the expression:

$$\phi = \frac{{}^{60}\text{Co activity}}{\lambda {}^{60}\text{N} {}^{\Delta 59}\text{T}}$$

λ^{60} = decay constant of 5.24-year ^{60}Co ,

N^{59} = number of ^{59}Co atoms in the monitor,

${}^{\Delta 59}$ = effective neutron capture cross section of ^{59}Co

= 36.9 barns for a reactor flux,

T = irradiation time.

MASS SPECTROMETRIC ANALYSES

The Mass Spectrometer

All the rare gas samples were analyzed in a 90° -sector ten-inch radius mass spectrometer with a resolving power of about 620. This proved sufficient to resolve rare gas isotopes from hydrocarbons at the same mass numbers. A schematic diagram of the instrument is shown in Figure 11.

Gas atoms or molecules are positively ionized in the source by electron bombardment and accelerated through a potential difference of about 2.5 Kv. A magnetic field deflects the ions and focuses them onto the detector slit. The detection is accomplished by an Allen-type electron multiplier with a gain of 10^5 . A vibrating reed electrometer amplifies the current output of the electron multiplier. The mass spectrum is scanned by slowly varying the magnetic field while maintaining the ion-accelerating voltage at a constant value. As the group of ions at each successive mass number passes across the detector slit, a peak results in the electrometer reading. A pen recorder traces these peaks onto a chart to form a permanent record of the analysis. A typical chart recording is shown in Figure 12.

Two mercury diffusion pumps, each with a liquid nitrogen trap, maintain a high vacuum inside the spectrometer. The diffusion pumps are backed by a rotary pump. When the spectrometer is not being used for analysis the pressure is about 5×10^{-9} mm. of mercury but may rise

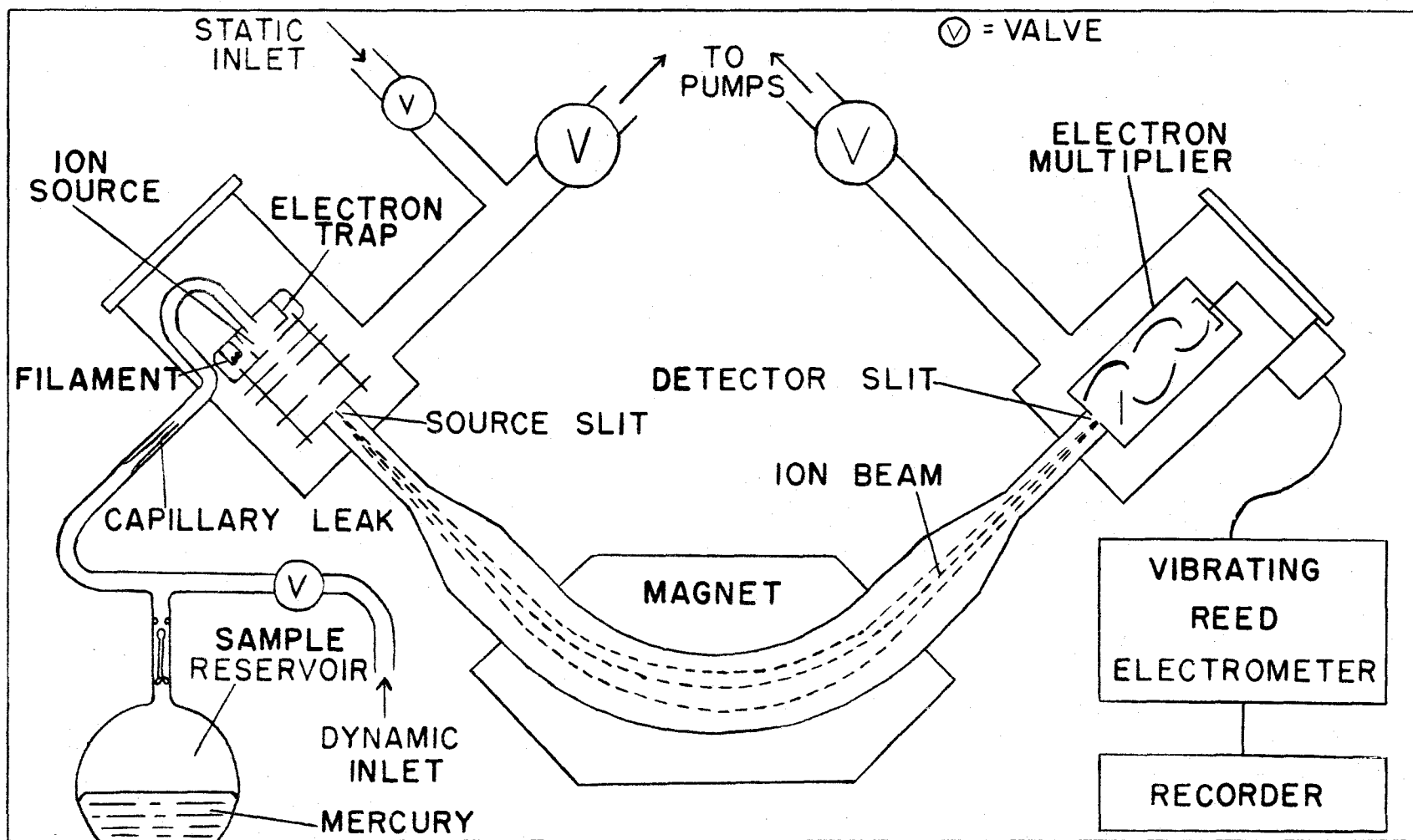


Fig. 11. Schematic diagram of the mass spectrometer.

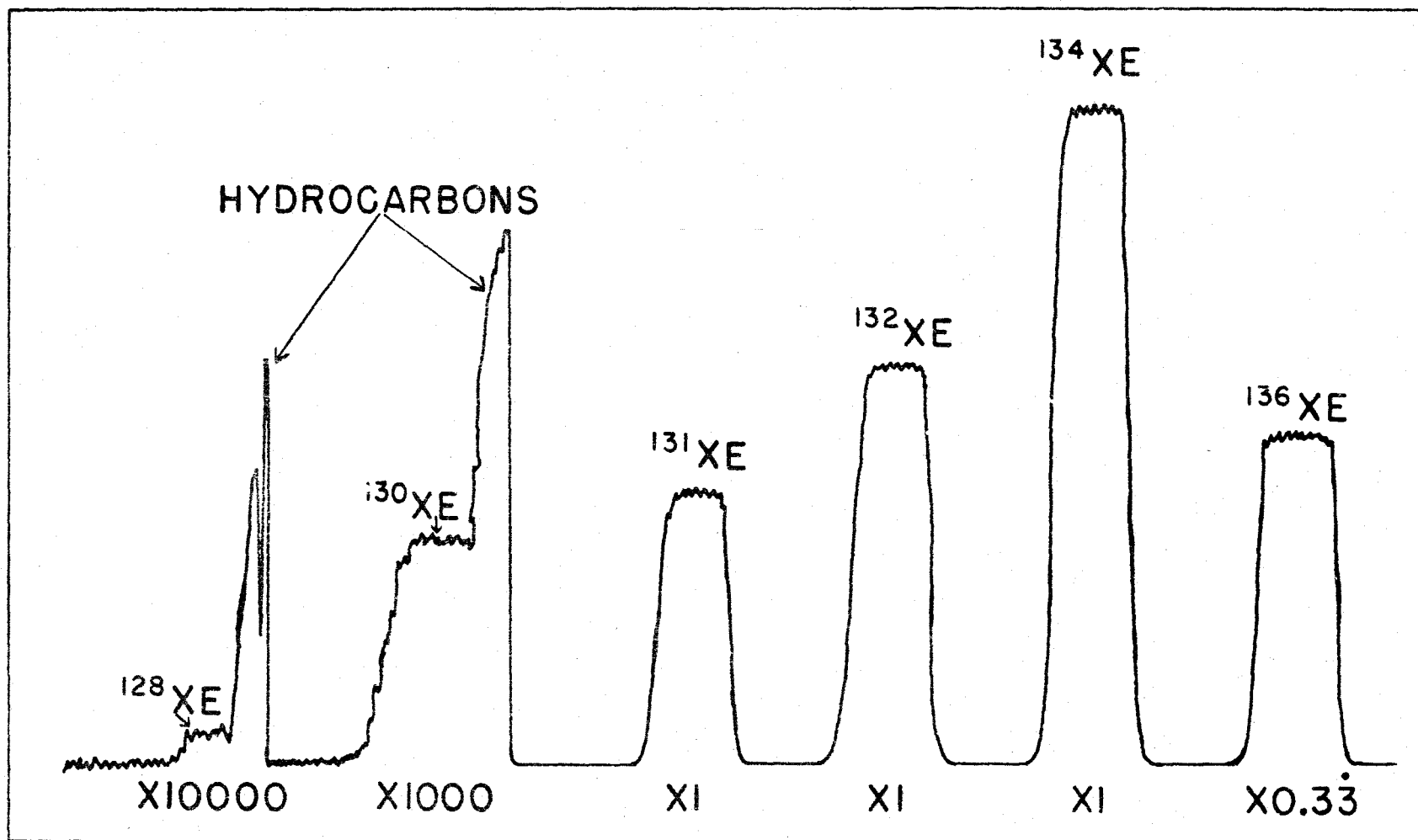


Fig. 12. Typical chart recording of the spectrum of fission product xenon.

to about 5×10^{-7} mm during an analysis.

Analyses of the Rare Gases

Gas samples may be analyzed either by the static or by the dynamic method. In the static method the valves to both pumps are closed and the entire sample is let into the mass spectrometer at once. This method is suited for the analysis of very small samples, where maximum sensitivity is required. Large gas samples cannot be analyzed in this way, because the pressure inside the spectrometer would rise too high and interfere with the flow of ions. This would lead to "pressure broadening" of the observed peaks.

Large samples are analyzed by the dynamic method, where only a small fraction of the sample is inside the spectrometer at any given time. The gas sample is allowed to leak slowly into the source area through a fine capillary tube. One or both of the pump valves are left open, and a dynamic equilibrium in the sample pressure is soon established. The flow rate through the capillary leak and thus the peak height is controlled by raising or lowering the mercury level in a sample reservoir behind the leak. This method is about 2000 times less sensitive than the static method.

The large rare gas samples intended for independent yield studies were analyzed by the dynamic method. Most of the smaller samples intended for cumulative yield measurements from neptunium fission were analyzed by the static method.

Analyses are performed by scanning up and down through the isotopes of each rare gas. The peak height for each isotope on the upward scan is added to its height on the downward scan. This is done in order to cancel any errors resulting from peak height attenuation due to sample loss. In a static analysis some of the sample is lost by becoming adsorbed on the walls of the spectrometer. In a dynamic analysis, the flow rate through the capillary leak decreases as the total sample behind the leak becomes depleted. Altogether, from six to ten double (upward and downward) scans are taken for each sample. Mass ratios are calculated for each double scan, and then averaged over all scans.

After each analysis, the remainder of the sample is recovered in a gas sample tube, removed from the mass spectrometer line, and stored for future reference.

Sources of Error

In addition to the possibility of previous contamination of the sample, the following errors may arise in the course of the mass spectrometric analysis itself.

1). Interference From Hydrocarbon Background

Even after the mass spectrometer has been baked extensively, hydrocarbons are still present at almost every mass number. The resolving power of the mass spectrometer, (620), is great enough to

separate completely a xenon or krypton peak if it is comparable in size to, or larger than, the adjacent hydrocarbon peak. However, if the rare gas peak is much smaller than the hydrocarbon, as in the case of the very low independent yields, then correction becomes difficult because the rare gas peak is partially obscured by the "tail" of the hydrocarbon peak. This effect is most serious in the case of ^{128}Xe and the accompanying hydrocarbon. For this reason, the independent ^{128}I yield measurements are not as precise as those of other independent yields.

2). Memory

Ions from a large sample tend to become embedded in the walls of the spectrometer and to evolve slowly, giving rise to a memory effect in subsequent analyses. This effect is most serious during a static analysis, when the evolving atoms accumulate in the spectrometer. In this case the effect is manifested as a systematic change in the isotopic ratios as a function of time. Flushing the spectrometer for several hours with nitrogen or neon usually eliminates the memory. If it persists, then the measured isotopic ratios are plotted as a function of time and the graphs extrapolated to zero time (the time when the sample is admitted). This gives the true isotopic ratios of the sample.

3). Mass Discrimination

Mass discrimination, i.e., unequal responses to the abundances of different isotopes of the same element, can arise in the capillary leak, the ion source, or the electron multiplier. Light isotopes diffuse through the capillary leak more rapidly than heavy isotopes, and after a period of time the sample in the reservoir behind the leak becomes depleted in the lighter isotopes. Analyses of atmospheric xenon and krypton show this effect to be negligible, provided not more than 10% of the sample has been expended.

The fact that the ion accelerating voltage is held constant while the magnetic field is scanned tends to minimize mass discrimination in the ion source. However, a small mass discrimination effect arises during static analyses. Analyses of atmospheric rare gases show a 1% enrichment in the ratio of ^{83}Kr to ^{86}Kr in comparison with Nier's (46) standard values. Thus a corresponding correction must be made to the measured fission yields. The effect is negligible in the xenon region.

RESULTS AND DISCUSSION

PART I. CUMULATIVE YIELDS FROM THE FISSION OF ^{237}Np AND ^{238}Np

The cumulative yields of the xenon and krypton isotopes from neptunium fission processes are recorded in Tables I to VI. The xenon yields are expressed relative to ^{132}Xe , and the krypton yields relative to ^{86}Kr . The isotope ^{132}Xe serves as a convenient basis in the xenon region because its yield generally does not deviate markedly from a smooth curve. Of the stable krypton isotopes, ^{86}Kr is produced with the greatest abundance in fission, and is therefore the least susceptible to the effects of atmospheric krypton contamination. Thus ^{86}Kr was chosen as a basis for the relative krypton yields.

The xenon yields have been corrected for atmospheric contamination whenever necessary from the abundance of ^{129}Xe . This isotope comprises 26.44% of atmospheric xenon, but its production in fission is blocked by the long-lived ^{129}I (half-life = 1.7×10^7 years). The krypton yields have been corrected for atmospheric contamination from the abundances of ^{82}Kr and ^{80}Kr , which comprise 11.56% and 2.27% respectively of atmospheric krypton. These isotopes of krypton are also produced from the independent yields of the shielded nuclides ^{82}Br and ^{80}Br , but the independent yields are negligible when compared with the cumulative yields of the other

krypton isotopes.

The error indicated for each yield in the following tables is the standard deviation of the individual isotopic ratio measurements about the mean value. It has been found in practice that this method of expressing the error best represents the reproducibility of the mean value of the isotopic ratio from one sample to the next.

The yields from the specific neptunium fission processes will now be discussed.

Yields from the Thermal Neutron Fission of ^{238}Np

The cumulative yields of the stable xenon isotopes from the thermal neutron fission of ^{238}Np are recorded in Table I, and the yields of the stable krypton isotopes in Table II. Table III shows the cumulative yields of various unstable isotopes -- ^{133}Xe , ^{135}Xe , ^{87}Kr , and ^{88}Kr -- from the fission of ^{238}Np .

In Table I the observed ^{136}Xe yields have been corrected for neutron capture on ^{135}Xe by using the expression derived in Appendix C. The irradiation times and neutron fluxes required for these corrections are recorded for each sample in Table I. The value used for the yield of ^{135}Xe is an average taken from Table III. Since the capture cross section of ^{135}Xe is extremely large (2.6×10^6 barns) and since relatively long irradiations in intense neutron fluxes are required to produce ^{238}Np fission, the capture corrections represent a significant portion of the observed ^{136}Xe yields. The larger errors

TABLE I

CUMULATIVE YIELDS OF THE STABLE XENON ISOTOPES FROM THE THERMAL NEUTRON FISSION OF ^{238}Np

Sample Number	Neutron Flux ($\text{n/cm}^2/\text{sec}$)	Irradiation Time (hours)	131/132	134/132	136/132
A-1	2×10^{14}	336	0.658 ± 0.004	1.616 ± 0.006	1.62 ± 0.05
A-2	2×10^{14}	336	0.668 ± 0.002	1.650 ± 0.006	1.66 ± 0.05
A-3	2×10^{14}	336	0.666 ± 0.002	1.635 ± 0.009	1.61 ± 0.05
A-4	1.21×10^{13}	720	0.686 ± 0.006	1.628 ± 0.005	1.75 ± 0.04
A-5	1.24×10^{13}	120	0.696 ± 0.006	1.607 ± 0.011	1.62 ± 0.02

TABLE II

CUMULATIVE YIELDS OF THE STABLE KRYPTON ISOTOPES
FROM THE THERMAL NEUTRON FISSION OF ^{238}Np

Sample Number	Neutron Flux (n/cm ² /sec)	Irradiation Time (hours)	83/86	84/86	85/86
A-1	2 x 10 ¹⁴	336	0.315 ± 0.002	0.558 ± 0.004	0.670 ± 0.003
A-2	2 x 10 ¹⁴	336	0.318 ± 0.003	0.562 ± 0.004	0.673 ± 0.005
A-3	2 x 10 ¹⁴	336	0.3219 ± 0.0006	0.566 ± 0.002	0.674 ± 0.003
A-5	1.24 x 10 ¹³	120	0.321 ± 0.004	0.544 ± 0.005	0.683 ± 0.006

TABLE III

CUMULATIVE YIELDS OF THE UNSTABLE XENON AND KRYPTON ISOTOPES
 FROM THE THERMAL NEUTRON FISSION OF ^{238}Np
 (^{133}Xe , ^{135}Xe , ^{87}Kr , and ^{88}Kr)

Sample Number		B-1	B-2	B-3
Irradiation Time (hours)		60.00	60.00	58.03
Irradiation to Extraction Time (hours)		8.30	7.63	4.25
Extraction to Analysis Time (hours)		5.75	4.33	2.03
Neutron Flux ($\text{n/cm}^2/\text{sec}$)		1.56×10^{13}	1.39×10^{13}	1.11×10^{13}
Xenon Yields	133/132	1.50 ± 0.04	1.492 ± 0.013	1.502 ± 0.014
	135/132	1.41 ± 0.02	1.340 ± 0.012	1.430 ± 0.014
	136/132	1.69 ± 0.03	1.69 ± 0.03	- - - - -
Krypton Yields	87/86	- - - - -	- - - - -	1.16 ± 0.06
	88/86	- - - - -	1.45 ± 0.02	1.35 ± 0.01

TABLE IV

CUMULATIVE YIELDS OF THE STABLE XENON AND KRYPTON ISOTOPES
FROM THE THERMAL NEUTRON FISSION OF ^{237}Np

Sample Number		C-1	C-2	C-3
Irradiation time (hours)		1680	720	720
Neutron Flux ($\text{n/cm}^2/\text{sec}$)		9.0×10^9	8.1×10^9	9.2×10^9
Xenon Yields	131/132	0.661 ± 0.012	0.678 ± 0.014	0.680 ± 0.006
	134/132	1.66 ± 0.02	1.65 ± 0.03	1.62 ± 0.04
	136/132	- - - - -	1.60 ± 0.02	1.57 ± 0.03
Krypton Yields	83/86	0.327 ± 0.014	- - - - -	- - - - -
	85/86	0.68 ± 0.02	- - - - -	- - - - -

TABLE V

CUMULATIVE YIELDS OF THE XENON AND KRYPTON ISOTOPES
FROM THE FAST NEUTRON FISSION OF ^{237}Np

Sample Number		D-1	D-2	D-3
Irradiation Time (hours)		4.00	4.00	2.00
Cadmium Shielded		yes	yes	no
Xenon Yields	131/132	0.746 ± 0.003	0.746 ± 0.002	0.732 ± 0.003
	133/132	- - - - -	- - - - -	1.322 ± 0.008
	134/132	1.472 ± 0.006	1.469 ± 0.006	1.464 ± 0.007
	136/132	1.369 ± 0.007	1.362 ± 0.010	1.382 ± 0.005
Krypton Yields	83/86	0.365 ± 0.006	0.362 ± 0.006	- - - - -
	84/86	0.53 ± 0.03	0.55 ± 0.03	- - - - -
	85/86	0.763 ± 0.013	0.764 ± 0.013	- - - - -

TABLE VI

SUMMARY OF THE XENON AND KRYPTON YIELDS
FROM THE VARIOUS NEPTUNIUM FISSION PROCESSES

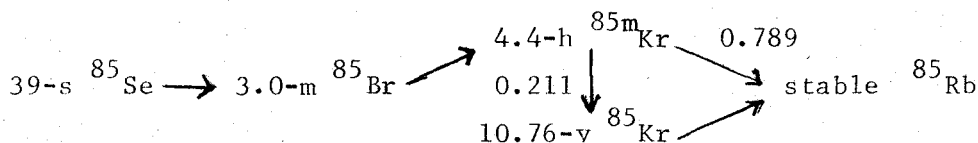
		^{238}Np Thermal neutron fission (from Tables I, II and III)	^{237}Np Thermal neutron fission (from Table IV)	^{237}Np Fast neutron fission (from Table V)
Xenon Yields	131/132	0.664 ± 0.004	0.673 ± 0.010	0.746 ± 0.003
	133/132	1.498 ± 0.014	- - - - -	1.32 ± 0.02
	134/132	1.634 ± 0.014	1.64 ± 0.03	1.470 ± 0.006
	135/132	1.39 ± 0.05	- - - - -	- - - - -
	136/132	1.65 ± 0.04	1.58 ± 0.03	1.366 ± 0.009
Krypton Yields	83/86	0.318 ± 0.003	0.327 ± 0.014	0.364 ± 0.006
	84/86	0.562 ± 0.004	- - - - -	0.54 ± 0.03
	85/86	0.672 ± 0.003	0.68 ± 0.02	0.763 ± 0.013
	87/86	1.16 ± 0.06	- - - - -	- - - - -
	88/86	1.40 ± 0.05	- - - - -	- - - - -

quoted for these yields result from uncertainties in this capture correction.

The 131/132 ratios from samples A-4 and A-5 are both somewhat high, and the 134/132 ratio from sample A-5 is low. These small shifts in isotopic ratios can be attributed to the effects of another fission process, probably the fission of ^{237}Np by fast neutrons, i.e., fission spectrum neutrons. The rate of fission of this nuclide depends linearly on the neutron flux, whereas the rate of ^{238}Np fission depends on the square of the neutron flux. Now for samples A-1, A-2, and A-3 the neutron fluxes are about a factor 15 greater than for samples A-4 and A-5. Thus the ratio of the number of ^{238}Np fissions to ^{237}Np fissions is 15 times as great for the first three samples as for the last two samples. This 15-fold increase in the proportion of ^{238}Np fissions leads to only very small shifts in the isotopic ratios. From this, we conclude that samples A-1, A-2, and A-3 represent pure ^{238}Np thermal neutron fission, whereas samples A-4 and A-5 represent a trace of another fission process in addition to that of ^{238}Np fission. This argument is equally valid whether this other process is indeed the fast neutron fission of ^{237}Np , or whether it is the result of any other nuclide whose fission rate is a linear function of flux. Thus the xenon yields from samples A-4 and A-5 are not included in the averages in Table VI.

The yield of ^{85}Kr has been included with the yields of the stable isotopes in Table II, because the ground state of this nuclide

has an appreciably long half-life (10.76 years). The fission product chain at mass 85 appears as follows:



The branching ratio of the decay of the isomeric state of ^{85}Kr to its ground state is taken from recent data by W. B. Clarke and H. Farrar (47). By the time the stable isotopes of krypton were analyzed, all of the 4.4-hour isomeric state of ^{85}Kr had decayed away. The total yield of ^{85}Kr was then determined from the abundance of the 10.76-year ground state, modified by a small correction for partial decay, and from the reported branching ratio of the isomer to the ground state.

In Table II the various isotopic ratios are consistent from sample to sample, except for sample A-5, which shows a small downward shift in the 84/86 ratio, and a small upward shift in the 85/86 ratio. These shifts can again be attributed to a small contribution from the fast neutron fission of ^{237}Np . Thus the krypton ratios from sample A-5 are omitted from the averages in Table VI.

The yields of the unstable isotopes of xenon and krypton must be measured shortly after irradiation, when the production of ^{132}Xe is still blocked by the incomplete decay of ^{132}Te (half-life = 77 hours) and the production of ^{131}Xe by the incomplete decay of

^{131}I (half-life = 8.05 days). For this reason the abundances of ^{131}Xe and ^{132}Xe were not measured for sample B-1, B-2, and B-3 in Table I. On the other hand, the yields of the 134 and 136 mass chains decay almost immediately to ^{134}Xe and ^{136}Xe respectively. Thus the yields of ^{133}Xe , ^{135}Xe , and ^{136}Xe for samples B-1, B-2, and B-3 were first measured relative to ^{134}Xe . These ratios were then converted to ^{132}Xe as a basis using the average 134/132 ratio for ^{238}Np fission in Table VI. The ^{136}Xe yields in Table III have been corrected for neutron capture as in Table I.

In the krypton region, the 86 mass chain decays almost immediately to ^{86}Kr . The yields of ^{87}Kr and ^{88}Kr were thus directly measured relative to ^{86}Kr . No corrections have been made for the effects of delayed neutron emission in the reported yields of ^{87}Kr and ^{88}Kr . However, these yields, as they stand, can be compared to the directly observed yields reported in the literature (e.g., Katcoff (23)) for other fissile nuclides. Actually, about three per cent of the mass 87 chain contributes to the observed ^{86}Kr yield through delayed neutron emission, while about seven per cent of the mass 88 chain contributes to the observed ^{87}Kr yield. Fifteen per cent of the mass 89 chain decays to ^{88}Kr by delayed neutron emission.

Corrections for the partial decay of the unstable xenon and krypton isotopes were carried out by means of the equations in Appendix B. The yield of ^{133}Xe (half-life = 5.27 days) can be determined quite accurately in this way. However, the yields of

^{135}Xe (half-life = 9.2 hours), ^{87}Kr (half-life = 76 minutes), and ^{88}Kr (half-life = 2.8 hours) are determined with considerably less accuracy. By the time the ^{87}Kr yield measurement was carried out for sample B-3, nearly all of the ^{87}Kr had decayed away. Hence the precision of this measurement is only about 5%. The quoted errors for the yields of ^{135}Xe and ^{88}Kr refer only to the precision of the isotopic ratio measurements on the mass spectrometer. Uncertainties in the half lives and decay times are mainly responsible for the differences in the observed yields of ^{135}Xe and ^{88}Kr from sample to sample.

The yields obtained from samples B-1, B-2, and B-3 may contain a small contribution from the fast neutron fission of ^{237}Np . It was seen in Table I that samples A-4 and A-5 showed a small shift of isotopic ratios due to this process. However, the uncertainties involved in measuring the yields of the unstable isotopes are undoubtedly greater than any small effects that might arise from the fission of ^{237}Np .

The average values of the various xenon and krypton yields from the thermal neutron fission of ^{238}Np are listed in the first column of Table VI. One of the most striking features of these results is the abnormally low yield of ^{135}Xe , compared to the yields of the adjacent isotopes ^{134}Xe and ^{136}Xe . It might be argued that the corrections made for neutron capture on ^{135}Xe were not large enough, perhaps because of incorrect information on the capture cross section of ^{135}Xe or on the neutron fluxes. In other words,

the ^{135}Xe might actually be higher, and the ^{136}Xe yield lower than the values reported in Table VI. This would lead to a smoother mass yield curve.

Such an argument, however, is refuted by a closer examination of the corrected ^{136}Xe yields from the individual samples in Tables I and III. Because of the high neutron fluxes used in the irradiations of sample A-1, A-2, and A-3, nearly 100% of the mass 135 yield contributes to the observed ^{136}Xe abundances. For samples A-4, A-5, B-1, and B-2, irradiated at lower fluxes, this contribution is only about half the mass 135 yield. Thus the neutron capture corrections are twice as great for the first three samples as for the remaining ones. If there were a systematic error in these corrections (e.g., a wrong value for the mass 135 yield), then there would be inconsistencies in the reported ^{136}Xe yields. As a matter of fact, the corrected ^{136}Xe yields from all the samples (except sample A-4) are consistent with one another within the quoted errors. Thus the reported low yield at mass 135 and the high yield at mass 136 appear to be real.

Yields from the Thermal Neutron Fission of ^{237}Np

The cumulative yields of the stable xenon and krypton isotopes (including ^{85}Kr) from the thermal neutron fission of ^{237}Np are recorded in Table IV. The very long irradiations required to produce a sufficient number of fissions at such a low neutron flux precluded

the determination of the yields of the unstable isotopes. The sample sizes were extremely small - less than 10^8 atoms at each isotope. For this reason the precision of these measurements is not as good as for those of ^{238}Np fission. Traces of contamination by atmospheric krypton precluded the determination of the krypton yields, except for the ^{83}Kr and ^{85}Kr yields from sample C-1. At these very low neutron fluxes, no capture corrections were necessary for the ^{136}Xe yield.

The average values of the yields from the thermal neutron fission of ^{237}Np are summarized in the second column of Table VI. From the remarkable similarity of these yields to the yields from ^{238}Np fission, it might be argued that the former yields are the result of ^{238}Np fission rather than ^{237}Np fission. This would occur if the reported cross section (0.019 barns), (48), for the thermal neutron fission of ^{237}Np were in error. However, calculations based on the accepted cross sections and on equation (1) on page 47 indicate that 97% of the total number of fissions in samples C-1, C-2, and C-3 result from ^{237}Np . The reported cross section for the thermal neutron fission of ^{237}Np would have to be in error by more than an order of magnitude in order for the yields from ^{238}Np fission to predominate in samples C-1, C-2, and C-3. Unless this cross section is found to be erroneous, the results in the second column of Table VI should be taken to represent the yields from the thermal neutron fission of ^{237}Np .

Yields from the Fast Neutron Fission of ^{237}Np

Table V shows the yields of the xenon and krypton isotopes from the fission of ^{237}Np by fast neutrons, i.e., fission spectrum neutrons. Samples D-1 and D-2 were wrapped in cadmium during irradiation, but sample D-3 was not. The yields of the stable xenon isotopes from this last sample are obviously affected to a small extent by thermal neutron fission and are therefore not included in the average yields of Table VI. It was possible, however, to obtain a yield for ^{133}Xe from sample D-3. Since this is the only measurement available for the yield of ^{133}Xe from the fast fission of ^{237}Np , it has been included in the summary in Table VI. The error quoted for this measurement in the latter table takes into consideration the possibility of thermal neutron fission in sample D-3.

Comparison of the Yields from ^{238}Np Fission with Those from Other Fissile Nuclides

It is of interest, first of all, to compare the yields from the thermal neutron fission of ^{238}Np with those from other fissile nuclides with large thermal fission cross sections. Figure 13 shows the absolute cumulative yields in the xenon region from three other fissile nuclides -- ^{235}U , ^{233}U , and ^{239}Pu . Although they are not shown, the absolute yields from ^{241}Pu fission have also been measured (8), and lie close to the ^{235}U yields in the mass range

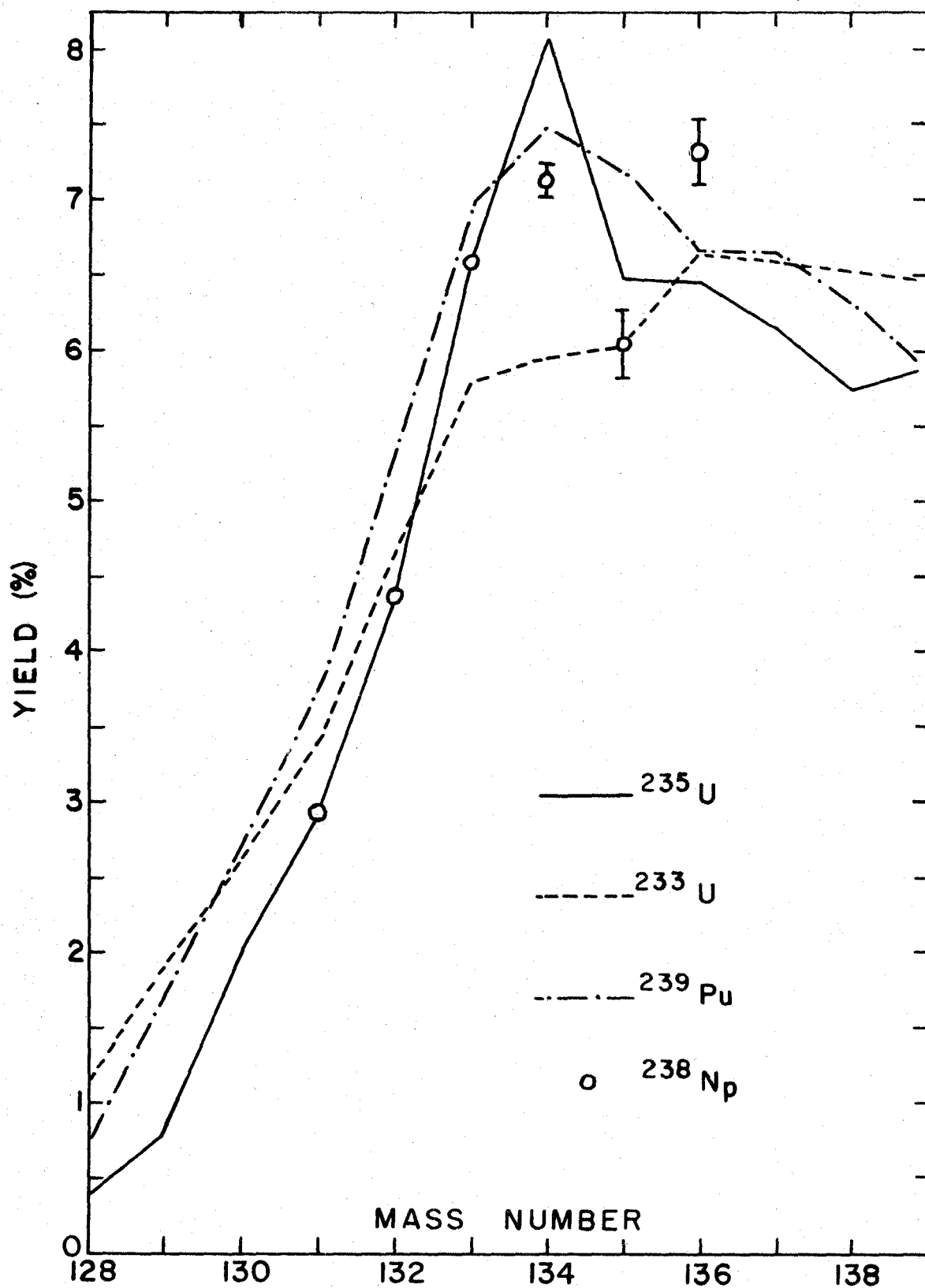


Fig. 13. Comparison of the cumulative xenon yields from ^{238}Np thermal neutron fission with the yields from other thermal neutron fission processes. The 132 yield from ^{238}Np has been arbitrarily set equal to the 132 yield from ^{235}U .

131 - 134. For the sake of comparison, the yield of ^{132}Xe from ^{238}Np fission is arbitrarily set equal to the ^{132}Xe yield from ^{235}U fission. The remaining xenon yields from ^{238}Np fission are then plotted in Figure 13 relative to this 132 yield.

In this mass region, the yields from ^{238}Np fission resemble those from ^{235}U fission more closely than the yields from the fission of ^{233}U or ^{239}Pu . In particular, the slope of the ^{238}Np yield curve from mass 131 to 133 is the same as that of the ^{235}U yields but is steeper than the slopes of the ^{233}U and ^{239}Pu yields. This variation in slope from one fissile nuclide to another can be correlated to some degree with the neutron-to-proton ratio of the compound fissioning nucleus. A higher neutron-to-proton ratio leads to a steeper curve. Table VII shows these ratios for the fissile nuclides whose yields have been studied in detail. The quantity $Y(132)/Y(131)$ is taken to represent the relative steepness of the mass yield curve in the range of 131 to 133. It is apparent that the slopes fall into two distinct groups. The nuclides with neutron-to-proton ratios of 1.559 or more have markedly steeper slopes than those with ratios less than 1.559.

As in the cases of ^{235}U and ^{241}Pu fission, we find pronounced fine structure in the ^{238}Np yields at mass 134. The ^{238}Np results also show a high yield at mass 136, a feature not observed in ^{235}U or ^{241}Pu fission. In fact the zig-zag variation in the ^{238}Np yields from mass 133 to 136 is quite unique.

TABLE VII

RELATIONSHIP BETWEEN NEUTRON-TO-PROTON RATIO OF FISSIONING NUCLEUS
AND SLOPE OF MASS YIELD CURVE

Fission process	Neutron-to-proton ratio of compound fissioning nucleus	$\frac{Y(132)}{Y(131)}$	Reference
$^{233}\text{U} + n$	1.544	1.37	10
$^{241}\text{Am} + n$	1.547	1.34	9
$^{239}\text{Pu} + n$	1.553	1.40	7
$^{242}\text{Am} + n$	1.558	1.38	9
$^{237}\text{Np} + n$	1.559	1.49	present work
$^{235}\text{U} + n$	1.565	1.49	6
$^{238}\text{Np} + n$	1.570	1.51	present work
$^{241}\text{Pu} + n$	1.575	1.48	8

It is generally observed that the position of the light mass peak shifts to progressively higher mass numbers as the mass of the fissioning nucleus increases. The krypton yields, lying on the lower side of the light mass peak, undergo a corresponding decrease. On this basis, one would expect the krypton yields from ^{238}Np fission to be lower than those from ^{235}U or ^{233}U fission, but higher than those from ^{239}Pu fission. Unfortunately, it is not possible to verify this prediction directly because the krypton yields from ^{238}Np are not known in an absolute sense, nor have they been determined relative to the heavy mass peak which remains more or less fixed from one fissile nuclide to another.

It is possible, however, to draw some conclusions by examining the slopes of the various krypton yield curves. Figure 14 shows the absolute krypton yields from the fission of ^{233}U , ^{235}U , and ^{239}Pu . These curves illustrate clearly the shift of the light mass peak to higher mass numbers. The yield of ^{86}Kr from ^{238}Np fission is then arbitrarily placed midway between the yields from the fission of ^{235}U and ^{239}Pu , and the other krypton yields from ^{238}Np are plotted relative to this point. When displayed in this way, the slope of the ^{238}Np yield curve is steeper than that from ^{239}Pu fission in the range of mass 83 to 86, but not as steep as the curve from ^{235}U fission. Also, the curve from ^{238}Np fission shows no tendency to cross the curves from the other fissile nuclides. Thus it appears that the position of the light mass peak from ^{238}Np

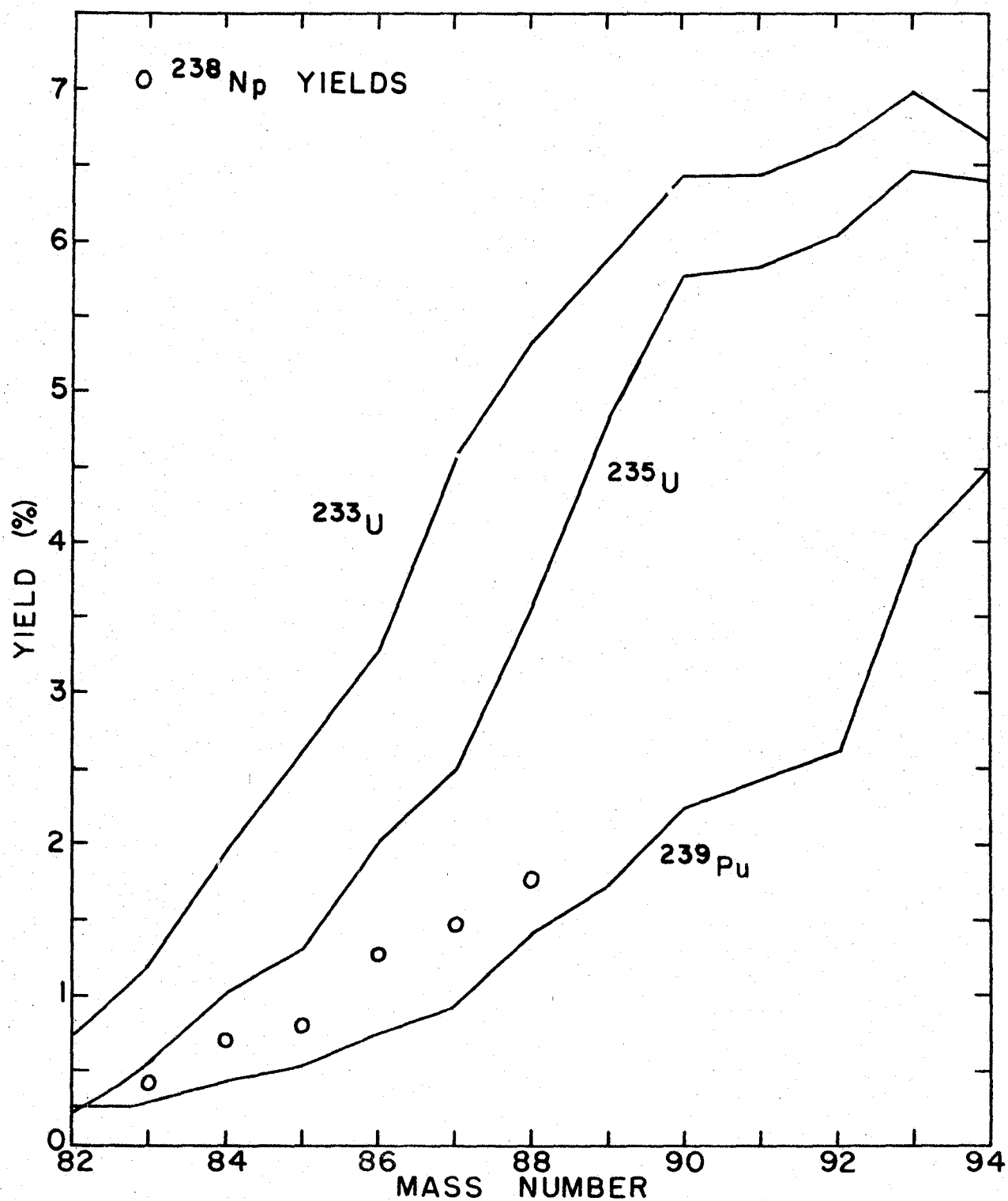


Fig. 14. Comparison of the cumulative krypton yields from ^{238}Np thermal neutron fission with the yields from other thermal neutron fission processes. The 86 yield from ^{238}Np has been arbitrarily placed midway between the ^{235}U and ^{239}Pu curves.

fission does lie between the positions of the light mass peaks from the fission of ^{235}U and ^{239}Pu , in accordance with the general trend.

As in other fission processes, the yields from the thermal neutron fission of ^{238}Np show deviations from a smooth curve in the krypton region. The 84 yield is somewhat high, and the 85 yield is low.

Comparison of the Yields from the Various Neptunium Fission Processes

The yields from the other neptunium fission processes are compared with those from the thermal neutron fission of ^{238}Np in Figure 15, where the ^{132}Xe yields and the ^{86}Kr yields are set equal to 1.000. There is very little difference between the xenon yields from the thermal neutron fission of ^{237}Np and ^{238}Np , although the high yield at mass 136 may be slightly lower for ^{237}Np thermal neutron fission than for ^{238}Np fission. Also, there are no significant differences in the krypton region between the yields from these two thermal fission processes.

On the other hand, the mass yield curve from the fast neutron fission of ^{237}Np is distinctly broader in the xenon region than the curve from the thermal neutron fission of ^{237}Np . A similar effect is observed in ^{235}U when comparing the yields from fission by thermal neutrons with the yields from fission by 14 Mev neutrons, (23). The fission spectrum neutrons used to study the fast neutron fission of ^{237}Np have an average energy of about one Mev. Apparently the fission

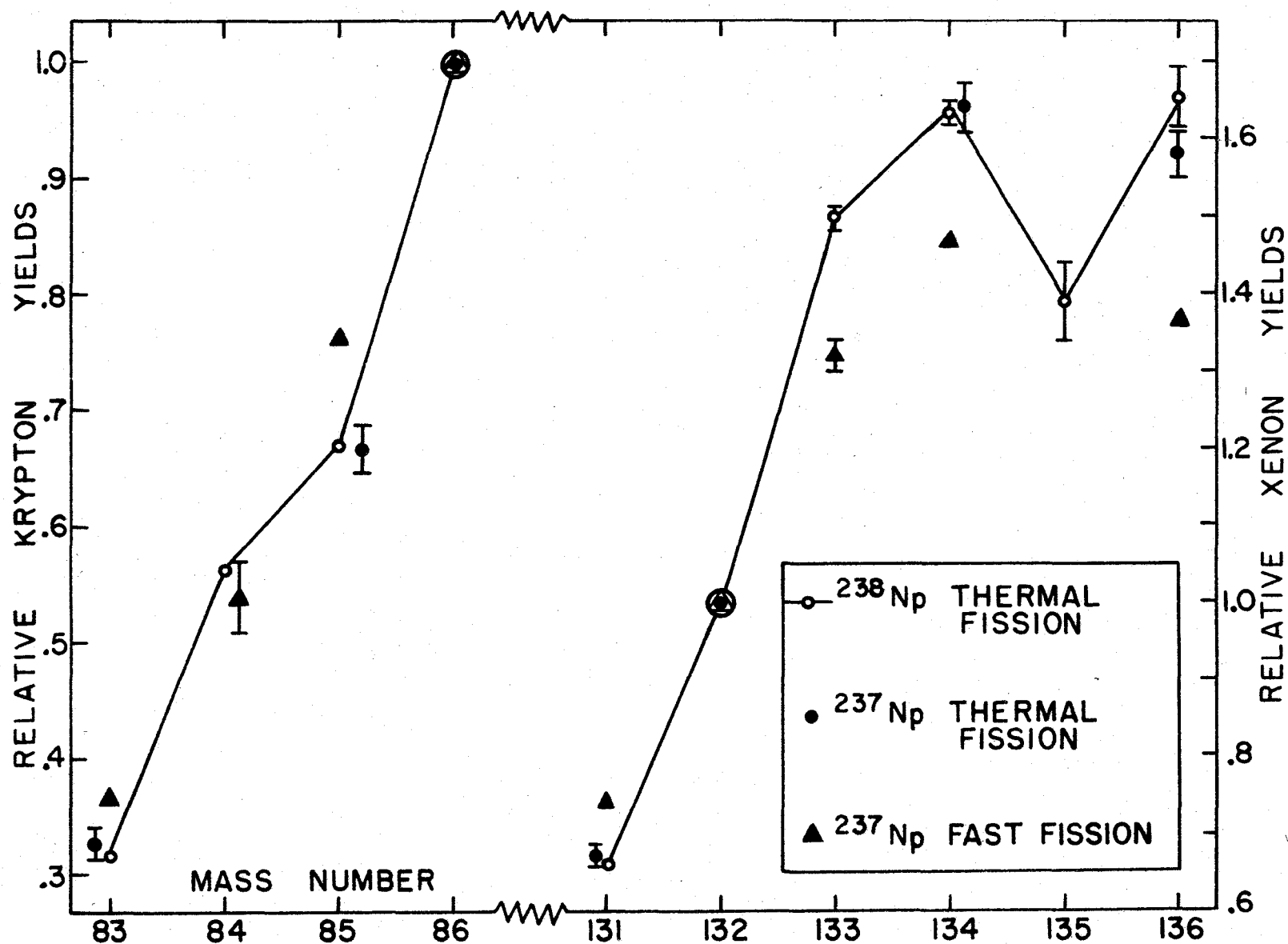


Fig. 15. Comparison of the cumulative krypton and xenon yields from the three fission processes in neptunium.

The xenon yields have been normalized at mass 132, and the krypton yields at mass 86.

process becomes less selective at higher neutron energies.

The present results on ^{237}Np fission also show that fine structure, so prominent at thermal neutron energies, almost completely disappears at higher neutron energies. For the fast neutron fission of ^{237}Np , the 136 yield is not abnormally high, and the mass yield curve in the krypton region is quite smooth. However, there is still some fine structure at mass 134.

The similarity in the yields from the thermal neutron fission of the two different isotopes of neptunium is surprising, especially when there are such marked differences between the yields from the fission of ^{233}U and ^{235}U , as shown in Figure 13, or between the yields from the fission of ^{239}Pu and ^{241}Pu . However, in these cases the neutron number increases by two in going from one isotope to the next. On the other hand, the neutron number increases by only one in going from ^{237}Np to ^{238}Np . Thus we might expect the differences in the yields from this set of isotopes to be less pronounced.

Pleva (9) observed that differences do occur between the xenon yields from the fission of ^{241}Am and ^{242}Am , although these differences are much less striking than for the isotopes of uranium or plutonium. Table VIII shows Pleva's results on the xenon and krypton yields from the thermal neutron fission of ^{241}Am and ^{242}Am . In this case the xenon yields were measured relative to ^{132}Xe , and the krypton yields relative to ^{84}Kr . The ^{136}Xe yield from ^{242}Am is

TABLE VIII

CUMULATIVE XENON AND KRYPTON YIELDS
 FROM THE THERMAL NEUTRON FISSION OF ^{241}Am AND ^{242}Am

		^{241}Am	^{242}Am
Xenon Yields	131/132	0.747 ± 0.003	0.725 ± 0.005
	133/132	1.15 ± 0.01	- - - - -
	134/132	1.52 ± 0.01	1.6 ± 0.1
	136/132	1.60 ± 0.03	1.9 ± 0.1
Krypton Yields	83/84	0.65 ± 0.04	0.65 ± 0.06
	85/84	1.13 ± 0.11	1.1 ± 0.2
	86/84	1.52 ± 0.18	1.5 ± 0.3

Results taken from the mass spectrometric data of Pleva (9).

unusually high. However, the measurement of this yield involved a large correction for neutron capture on ^{135}Xe . The yield of the latter nuclide was not measured mass spectrometrically, but was taken from a radiochemical determination of much poorer precision. Thus it is doubtful whether the ^{136}Xe yield from this process is actually so high. There are no significant differences in the krypton yields from the fission of the two americium isotopes.

The results from neptunium and americium show that a change of one in the neutron number of the fissioning nucleus does not produce a large change in the mass yield curve.

Theoretical Implications

These results have an important bearing on the theory of fission. The asymmetric transition state model assumes that, in low energy fission, the main features of the fission process are governed by a relatively few nuclear states at the saddle point of the compound fissioning nucleus. If this is so, then the specific nature of the transition state spectrum should have an important influence on the shape of the mass yield curve.

Now the thermal neutron fission of most of the fissile nuclides whose yields have been studied in detail, i.e., ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu , proceeds through compound nuclei which are even-even with respect to proton and neutron numbers. The transition state spectra of such nuclei should all be quite similar.

On the other hand, the thermal neutron fission of ^{238}Np proceeds through an odd-even compound nucleus. Such a nucleus should have a considerably different transition state spectrum. The thermal neutron fission of ^{237}Np proceeds through an odd-odd compound nucleus, which should have yet a third type of spectrum. If the transition state spectra have an important influence on the mass distribution, then one would expect the mass yields from these three types of compound fissioning nuclei to show marked differences. However, we have seen that the yields from ^{238}Np fission are remarkably similar to those from the fission of ^{235}U or ^{241}Pu . Furthermore, the yields from the thermal neutron fission of ^{237}Np are almost identical to those from ^{238}Np fission and the yields from the fission of ^{241}Am and ^{242}Am are also quite similar. Apparently, in thermal neutron fission at least, the features of the mass yield curve are not determined by the particular spectrum of transition states, but rather by gross features in the fissioning nucleus that are quite similar from one nuclide to another in the region of mass 234 to 243.

The fine structure in the mass yield curve from ^{238}Np thermal neutron fission, particularly the zig-zag variation from mass 133 to 136, probably results from fluctuations in the neutron emission characteristics of the prompt fission fragments. At the present time it is not possible to make quantitative predictions about the degree of this fine structure because the prompt yields themselves are not

known for ^{238}Np fission, nor is the neutron emission mechanism fully understood. However, in the following section, some calculations of independent ^{128}I and ^{130}I yields are made on the basis of a simple theory of neutron emission.

PART II. INDEPENDENT YIELDS OF ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I

Method of Computation

The fission product mass chains of interest in the region of the independent bromine yields are shown in Figure 16, and those in the region of the independent iodine yields in Figure 17. The mass spectrometric measurements give the abundances of ^{80}Kr and ^{82}Kr relative to ^{83}Kr , and of ^{128}Xe and ^{130}Xe relative to ^{131}Xe . The absolute cumulative yields of ^{83}Kr and ^{131}Xe are well known for most fissile nuclides. However, before the measured isotopic ratios can be converted to the independent yields of the shielded bromine and iodine isotopes, corrections must be made for neutron capture on the end products of lower mass chains. For example, the yield of the 129 chain accumulates as ^{129}I (half-life = 1.7×10^7 years), which subsequently captures neutrons during irradiation to form ^{130}I . Since the independent yield of ^{130}I is very low compared to the cumulative yield of the 129 chain, the neutron capture contribution can be a sizable fraction of the total ^{130}I produced. Similarly, ^{81}Br and ^{127}I are the end products of their respective mass chains, and capture neutrons to form ^{82}Br and ^{128}I . On the other hand, the production of ^{79}Br in fission is blocked by the long-lived ^{79}Se (half-life = 6×10^4 years), and the neutron capture to form ^{80}Br need not be considered. The capture effect is greatest for the

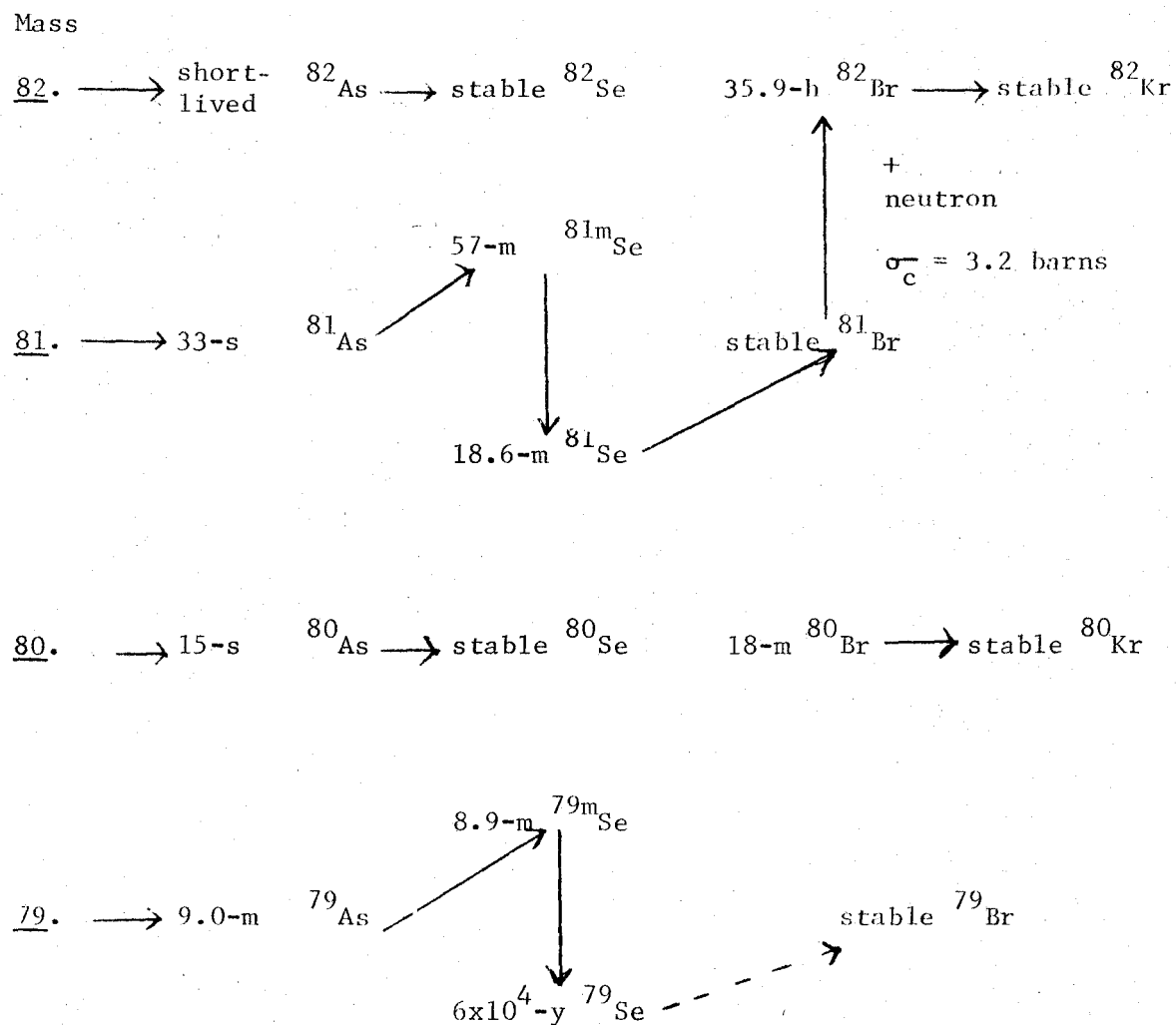


Fig. 16. Fission product mass chains in the region of the independent ^{82}Br and ^{80}Br yields.

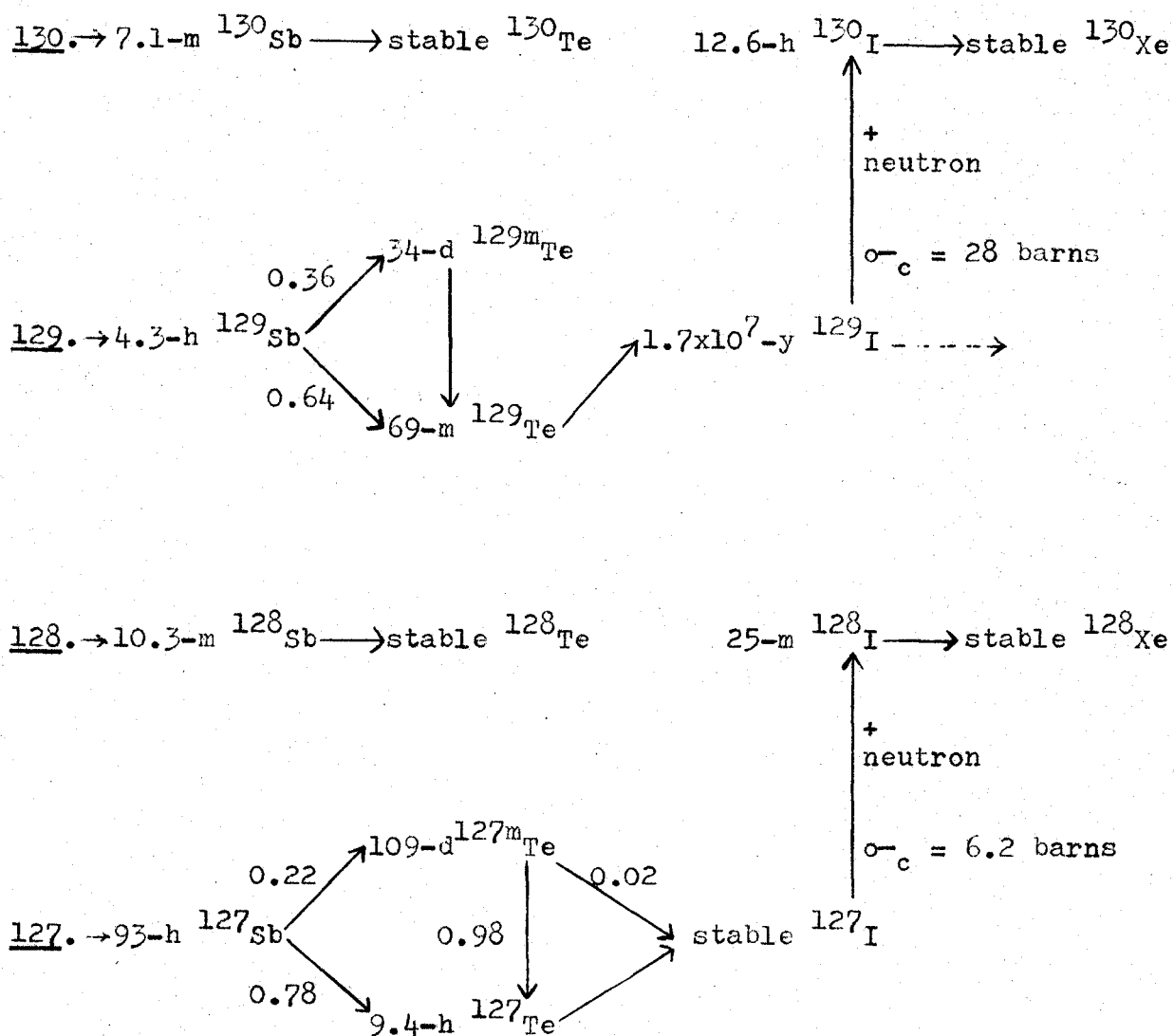
Mass

Fig. 17. Mass chains in the region of the independent ^{130}I and ^{128}I yields.

^{130}I yield, because of the comparatively high yield and capture cross section of ^{129}I . One can correct for these neutron capture effects, however, with a knowledge of the neutron flux, irradiation time, capture cross sections, and cumulative yields of the 81, 127, and 129 mass chains.

With the above considerations in mind, the absolute independent yields of the shielded nuclides ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I are given by the following expressions:

$$Y(^{80}\text{Br}) = \left[^{80}\text{Kr}/^{83}\text{Kr} \right] Y(83) \dots\dots\dots(2)$$

$$Y(^{82}\text{Br}) = \left[^{82}\text{Kr}/^{83}\text{Kr} \right] Y(83) - Y(81)\sigma_c^{81} \phi T/2 \dots\dots\dots(3)$$

$$Y(^{128}\text{I}) = \left[^{128}\text{Xe}/^{131}\text{Xe} \right] Y(131) - Y(127)\sigma_c^{127} \phi \int^{127}(T) \dots\dots\dots(4)$$

$$Y(^{130}\text{I}) = \left[^{130}\text{Xe}/^{131}\text{Xe} \right] Y(131) - Y(129)\sigma_c^{129} \phi \int^{129}(T) \dots\dots\dots(5)$$

The isotopic ratios $\left[^{80}\text{Kr}/^{83}\text{Kr} \right]$, $\left[^{82}\text{Kr}/^{83}\text{Kr} \right]$, $\left[^{128}\text{Xe}/^{131}\text{Xe} \right]$, and $\left[^{130}\text{Xe}/^{131}\text{Xe} \right]$ are measured by means of the mass spectrometer.

$Y(81)$, $Y(83)$, $Y(127)$, $Y(129)$, and $Y(131)$ are the absolute cumulative yields of the mass chains indicated in brackets. The accepted values of these cumulative yields for the fission of ^{235}U , ^{233}U , and ^{239}Pu are given in Table IX. σ_c^{81} , σ_c^{127} , and σ_c^{129} are the neutron capture cross sections of ^{81}Br , ^{127}I , and ^{129}I respectively. The

TABLE IX

SUMMARY OF CUMULATIVE YIELD DATA
USED IN THE COMPUTATION OF INDEPENDENT YIELDS

Mass Number	^{235}U	^{233}U	^{239}Pu
80	0.094%	0.20%	0.048%
81	0.14 %	0.40%	0.157%
82	0.24 %	0.75%	0.16 %
83	0.544%	1.17%	0.30 %
126	0.032%	- - -	- - -
127	0.13 %	0.60%	0.48 %
128	0.375%	1.1 %	0.71 %
129	0.8 %	1.9 %	1.7 %
130	2.0 %	2.6 %	2.7 %
131	2.93 %	3.39%	3.78 %

^{235}U and ^{239}Pu data are taken from the compilation by Meek and Ryder (49). ^{233}U data are taken from Katcoff's compilation (23).

values of these cross sections are shown in Figures 16 and 17.

ϕ is the average neutron flux, and $\mathcal{F}^{127}(T)$ and $\mathcal{F}^{129}(T)$ are functions of the irradiation time T . These functions take into consideration the incomplete β -decay of the precursors of ^{127}I and ^{129}I . The expressions for $\mathcal{F}^{127}(T)$ and $\mathcal{F}^{129}(T)$ are given in Appendix D.

In some cases it has been necessary to correct the observed isotopic ratios for traces of atmospheric contamination. Corrections to the observed ^{128}Xe and ^{130}Xe abundances were made from the abundance of ^{129}Xe . This isotope comprises 26.44% of atmospheric xenon, but its production in fission is blocked by the long-lived ^{129}I (half-life = 1.7×10^7 years).

Atmospheric corrections to the ^{80}Kr and ^{82}Kr abundances are more difficult to make. Because of the extremely low independent yield of ^{80}Br , the resulting ^{80}Kr is often completely obscured by atmospheric krypton contamination. In such cases only an upper limit can be set for the independent yield of ^{80}Br . The situation is less serious with the ^{82}Kr abundance, because the independent yields of ^{82}Br are generally from 10 to 100 times as great as the ^{80}Br yields. Thus, if the ^{80}Kr abundance is not unusually high, it can be concluded that the ^{82}Kr abundance is not seriously affected by atmospheric contamination.

TABLE X

 ^{235}U FISSION; INDEPENDENT YIELDS OF ^{130}I AND ^{128}I

Sample Number	Neutron Flux ($\times 10^{13} \text{ n/cm}^2/\text{sec}$)	Irradiation Time (hours)	$^{130}\text{Xe}/^{131}\text{Xe}$ $\times 10^{-4}$	Y (^{130}I) $\times 10^{-4} \%$	$^{128}\text{Xe}/^{131}\text{Xe}$ $\times 10^{-5}$	Y (^{128}I) $\times 10^{-5} \%$
E-1	1.97 ± 0.08	1450	- - - - -	- - - - -	1.38 ± 0.04	1.32 ± 0.16
E-2	1.17 ± 0.04	2137	- - - - -	- - - - -	1.32 ± 0.03	1.30 ± 0.13
E-3	1.7 ± 0.3	1632	- - - - -	- - - - -	1.55 ± 0.04	1.8 ± 0.5
E-5	1.2 ± 0.2	480	1.28 ± 0.01	2.2 ± 0.2	- - - - -	- - - - -
E-6	1.2 ± 0.2	1000	2.05 ± 0.03	2.6 ± 0.5	0.72 ± 0.06	1.08 ± 0.25
E-7	0.9 ± 0.2	333	1.05 ± 0.13	2.3 ± 0.4	- - - - -	- - - - -
E-8	0.9 ± 0.2	333	0.97 ± 0.01	2.1 ± 0.2	- - - - -	- - - - -
Average Independent Yields				(2.3 ± 0.3) $\times 10^{-4} \%$		(1.4 ± 0.3) $\times 10^{-5} \%$

TABLE XI

 ^{235}U FISSION; INDEPENDENT YIELDS OF ^{82}Br and ^{80}Br

Sample Number	Neutron Flux ($\times 10^{13} \text{ n/cm}^2/\text{sec}$)	Irradiation Time (hours)	$^{82}\text{Kr}/^{83}\text{Kr}$ $\times 10^{-4}$	Y (^{82}Br) $\times 10^{-5} \%$	$^{80}\text{Kr}/^{83}\text{Kr}$	Y (^{80}Br)
E-1	1.97 ± 0.08	1450	1.15 ± 0.10	4.0 ± 0.5	4.1×10^{-6}	$2 \times 10^{-6} \%$
E-2	1.17 ± 0.04	2137	1.28 ± 0.03	4.94 ± 0.17	9.5×10^{-7}	$5.2 \times 10^{-7} \%$
E-3	1.7 ± 0.3	1632	1.21 ± 0.05	4.2 ± 0.5	1.2×10^{-6}	$7.0 \times 10^{-7} \%$
E-4	1.7 ± 0.3	1099	1.27 ± 0.08	5.3 ± 0.5	4.5×10^{-6}	$2 \times 10^{-6} \%$
E-6	1.2 ± 0.2	1000	~ 1	~ 5	- - - - -	- - - - -
Average Independent Yields				(4.6 ± 0.5) $\times 10^{-5} \%$		(6 ± 1) $\times 10^{-7} \%$

TABLE XII

^{233}U FISSION; INDEPENDENT YIELDS OF ^{130}I AND ^{128}I

Sample Number	Neutron Flux ($\times 10^{13}$ n/cm ² /sec)	Irradiation Time (hours)	$^{130}\text{Xe}/^{131}\text{Xe}$ ($\times 10^{-4}$)	Y (^{130}I) ($\times 10^{-3}$ %)	$^{128}\text{Xe}/^{131}\text{Xe}$ ($\times 10^{-5}$)	Y (^{128}I) ($\times 10^{-5}$ %)
F-1	1.2 ± 0.3	250	8.31 ± 0.04	2.64 ± 0.05	1.26 ± 0.15	3.7 ± 0.5
F-2	1.2 ± 0.3	250	9.08 ± 0.30	2.90 ± 0.11	1.11 ± 0.23	3.2 ± 0.8
F-3	1.2 ± 0.3	250	9.38 ± 0.20	3.00 ± 0.08	1.09 ± 0.07	3.1 ± 0.3
F-4	0.9 ± 0.3	360	9.70 ± 0.16	3.09 ± 0.08	- - - - -	- - - - -
Average Independent Yields				(2.91 ± 0.17) $\times 10^{-3}$ %		(3.3 ± 0.5) $\times 10^{-5}$ %

TABLE XIII

 ^{233}U FISSION; INDEPENDENT YIELDS OF ^{82}Br and ^{80}Br

Sample Number	Neutron Flux ($\times 10^{13}$ n/cm ² /sec)	Irradiation Time (hours)	$^{82}\text{Kr}/^{83}\text{Kr}$ ($\times 10^{-3}$)	Y (^{82}Br) ($\times 10^{-3}$ %)	$^{80}\text{Kr}/^{83}\text{Kr}$	Y (^{80}Br)
F-1	1.2 ± 0.3	250	1.10 ± 0.11	1.28 ± 0.13	$< 2 \times 10^{-5}$	$< 2 \times 10^{-5}\%$
F-2	1.2 ± 0.3	250	0.82 ± 0.06	0.95 ± 0.07	$< 2 \times 10^{-5}$	$< 2 \times 10^{-5}\%$
F-3	1.2 ± 0.3	250	1.13 ± 0.27	1.3 ± 0.3	- - - - -	- - - - -
F-4	0.9 ± 0.3	360	1.10 ± 0.03	1.28 ± 0.03	$< 2 \times 10^{-5}$	$< 2 \times 10^{-5}\%$
Average Independent Yields				(1.20 ± 0.15) $\times 10^{-3}\%$		$< 2 \times 10^{-5}\%$

TABLE XIV

 ^{239}Pu FISSION; INDEPENDENT YIELDS OF ^{130}I AND ^{128}I

Sample Number	Neutron Flux ($\times 10^{13} \text{ n/cm}^2/\text{sec}$)	Irradiation Time (hours)	$^{130}\text{Xe}/^{131}\text{Xe}$ $\times 10^{-3}$	Y (^{130}I) $\times 10^{-3} \%$	$^{128}\text{Xe}/^{131}\text{Xe}$ $\times 10^{-3}$	Y (^{128}I) $\times 10^{-5} \%$
G-1	1.0 ± 0.3	224	1.42 ± 0.03	5.26 ± 0.11	- - - - -	- - - - -
G-2	0.70 ± 0.20	336	1.40 ± 0.01	5.16 ± 0.06	2.82 ± 0.10	10.3 ± 0.4
G-3	1.3 ± 0.4	224	1.40 ± 0.03	5.15 ± 0.12	2.1 ± 0.5	7.4 ± 1.9
G-4	1.05 ± 0.04	576	1.390 ± 0.004	4.90 ± 0.02	2.68 ± 0.10	8.6 ± 0.4
G-5	1.15 ± 0.04	672	1.392 ± 0.011	4.81 ± 0.04	2.80 ± 0.15	8.5 ± 0.5
Average Independent Yields				(5.06 ± 0.17) $\times 10^{-3} \%$		(8.7 ± 1.0) $\times 10^{-5} \%$

TABLE XV

^{239}Pu FISSION; INDEPENDENT YIELDS OF ^{82}Br and ^{80}Br

Sample Number	Neutron Flux ($\times 10^{13} \text{ n/cm}^2/\text{sec}$)	Irradiation Time (hours)	$^{82}\text{Kr}/^{83}\text{Kr}$ $\times 10^{-4}$	Y (^{82}Br) $\times 10^{-4} \%$	$^{80}\text{Kr}/^{83}\text{Kr}$	Y (^{80}Br)
G-1	1.0 ± 0.3	224	5.6 ± 0.9	1.66 ± 0.27	$\leq 1 \times 10^{-5}$	$\leq 3 \times 10^{-6} \%$
G-2	0.70 ± 0.20	336	4.8 ± 0.3	1.42 ± 0.08	$\leq 5 \times 10^{-6}$	$\leq 1.5 \times 10^{-6} \%$
G-3	1.3 ± 0.4	224	4.9 ± 1.4	1.5 ± 0.4	- - - -	- - - -
G-4	1.05 ± 0.04	576	6.8 ± 0.5	1.49 ± 0.15	$\leq 2 \times 10^{-5}$	$\leq 6 \times 10^{-6} \%$
Average Independent Yields				(1.52 ± 0.18) $\times 10^{-4} \%$		$\leq 1.5 \times 10^{-6} \%$

TABLE XVI

^{238}Np FISSION; FRACTIONAL CHAIN YIELDS OF ^{130}I , ^{128}I , and ^{82}Br

Sample Number	A-1	A-2	A-3	Average fractional chain yield
$^{130}\text{Xe}/^{131}\text{Xe} \times 10^{-4}$	- -	8.31 ± 0.04	9.03 ± 0.10	- - - - -
$F(^{130}\text{I}) \times 10^{-4}$	- -	4.8 ± 1.8	5.9 ± 1.8	$(5.4 \pm 1.8) \times 10^{-4}$
$^{128}\text{Xe}/^{131}\text{Xe} \times 10^{-5}$	- -	4.7 ± 0.7	3.3 ± 0.2	- - - - -
$F(^{128}\text{I}) \times 10^{-4}$	- -	3.0 ± 0.5	1.8 ± 0.2	$\leq (2.4 \pm 0.6) \times 10^{-4}$
$^{82}\text{Kr}/^{83}\text{Kr}$	$\leq 7 \times 10^{-4}$	- - - - -	- - - - -	- - - - -
$F(^{82}\text{Br})$	$\leq 1.4 \times 10^{-3}$	- - - - -	- - - - -	$\leq 1.4 \times 10^{-3}$

Each sample was irradiated for 336 hours at a flux of 2×10^{14} n/cm²/sec.

Yields from ^{235}U Fission

The absolute independent yields of ^{130}I and ^{128}I from the thermal neutron fission of ^{235}U are given in Table X, and those of ^{82}Br and ^{80}Br from the same fissioning nuclide are given in Table XI. The second and third columns in each table give the average neutron fluxes and irradiation times for each sample. The fourth and sixth columns give the measured isotope ratios after corrections have been made for any contamination by atmospheric rare gases or naturally occurring halogens. The errors quoted here are the standard deviations of the distributions of individual peak height ratios. The fifth and seventh columns give the computed absolute independent yields of the halogen nuclides. The errors in these columns include the errors in the neutron capture corrections and in the measured isotopic ratios.

In all, eight ^{235}U samples were analyzed. Samples E-1, E-2, E-3, and E-4 were purified by induction heating, and were believed to have a higher degree of purity. Also, these samples were irradiated for longer times at higher fluxes, and therefore gave more precise data on the very low independent yields of ^{128}I , ^{82}Br , and ^{80}Br . The neutron capture corrections for ^{130}I from these samples, however, were so large that they obscured the independent yield of ^{130}I . Samples E-5, E-6, E-7, and E-8 were irradiated for shorter times at lower fluxes, and gave more precise data for the ^{130}I yield.

The ^{128}I yield value from sample E-6 agrees with the values from the other samples within the limits of experimental error. Since sample E-6 was purified by a different method than samples E-1, E-2, and E-3, the possibility of the observed ^{128}I yields being seriously affected by natural iodine contamination is remote. The major portion of the errors in the ^{130}I and ^{128}I yields arises from uncertainties in the neutron capture corrections rather than in the measured isotopic ratios.

All the independent yield measurements are consistent with one another except for those of ^{80}Br . In Table XI it is seen that the $^{80}\text{Kr}/^{83}\text{Kr}$ values are much higher in samples E-1 and E-4 than in samples E-2 and E-3. The higher values are apparently due to contamination. The lower values are taken to represent the independent yield of ^{80}Br . Even in these cases, however, the possibility of atmospheric contamination cannot be entirely ruled out. For each sample in Table XI, the observed ^{80}Kr abundance has been multiplied by the ratio of ^{82}Kr to ^{80}Kr in the atmosphere (11.56/2.27) in order to give the possible atmospheric contribution to the observed ^{82}Kr abundance. This possible contribution has been included in the uncertainty of the $^{82}\text{Kr}/^{83}\text{Kr}$ measurement.

If in a given case all the observed ^{80}Kr were due to neutron capture on natural bromine, then the contribution of such contamination to the ^{82}Kr abundance would be negligible, since a neutron irradiation of natural bromine produces only one third as much ^{82}Kr as ^{80}Kr .

The averages of the various independent yields are shown at the bottom of the tables.

^{126}I : In samples E-1 and E-2 a search was made for ^{126}Xe resulting from the independent yield of the shielded nuclide ^{126}I . No ^{126}Xe was observed, and a upper limit of $1 \times 10^{-6}\%$ was established for the independent yield of ^{126}I .

Yields from ^{233}U Fission

The absolute independent yields from the thermal neutron fission of ^{233}U are shown in Table XII (^{130}I and ^{128}I yields) and Table XIII (^{82}Br and ^{80}Br yields). The tables are constructed in a manner similar to those for ^{235}U fission. The ^{80}Kr peak was not observed, and it was possible only to set an upper limit for the independent yield of ^{80}Br . This upper limit is still two orders of magnitude lower than the measured yield of ^{82}Br . Thus there can be no significant natural contamination contributing to the ^{82}Br yield. The ^{130}I , ^{128}I and ^{82}Br independent yield values are consistent for the four samples analyzed. Averages are taken and shown at the bottoms of the tables.

Yields from ^{239}Pu Fission

The absolute independent yields of ^{130}I and ^{128}I from the thermal neutron fission of ^{239}Pu are shown in Table XIV. The independent ^{82}Br

and ^{80}Br yields from ^{239}Pu are shown in Table XV. The tables are constructed in a manner similar to those for ^{235}U fission. In Table XIV, it is noted that the spread in absolute yield values is much greater than the spread in isotopic ratios. This increase in spread can be attributed to uncertainties in the corrections for neutron capture processes. The independent iodine yield values are averaged, and the spread is represented as a standard deviation.

In the krypton region (Table XV) only an upper limit could be set for the independent ^{80}Br yield. This is still two orders of magnitude lower than the measured ^{82}Br yield. Thus there can be no significant natural contamination contributing to the ^{82}Br yield. The values for this yield are consistent from sample to sample, and are averaged.

Yields from ^{238}Np Fission

Since none of the absolute cumulative yields for ^{238}Np fission are known, it was not possible to determine absolute independent yields for this nuclide. However, it is the fractional chain yields that are most useful in discussing theories of charge distribution in fission. It is possible to derive information about the fractional chain yields of ^{82}Br , ^{128}I , and ^{130}I from the measured ratios $\left[^{82}\text{Kr}/^{83}\text{Kr}\right]$, $\left[^{128}\text{Xe}/^{131}\text{Xe}\right]$, and $\left[^{130}\text{Xe}/^{131}\text{Xe}\right]$.

It was seen in Figure 13 of the previous section that the mass yield curve for ^{238}Np fission has the same shape as that for ^{235}U

fission in the mass range 131 to 133. Although no yields were measured for ^{238}Np fission in the mass range 127 to 131, it is not an unreasonable assumption that the mass yield curves from the two fission processes have the same shape in this region also. Mass yield curves in this region are generally quite smooth, and their shapes are not drastically different from one fissile nuclide to another. Thus, to a first approximation at least, the yields of the mass chains 127, 128, 129, and 130 expressed relative to that of 131 are the same for ^{238}Np fission as for ^{235}U fission. On this basis the fractional chain yields of ^{128}I and ^{130}I can be expressed as follows:

$$F(^{128}\text{I}) = \left[^{128}\text{Xe}/^{131}\text{Xe} \right] \left[Y(131)/Y(128) \right] - \left[Y(127)/Y(128) \right] \\ \times \sigma_c^{127} \phi \mathcal{F}^{127}(\text{T}) \dots \dots \dots (6)$$

$$F(^{130}\text{I}) = \left[^{130}\text{Xe}/^{131}\text{Xe} \right] \left[Y(131)/Y(130) \right] - \left[Y(129)/Y(130) \right] \\ \times \sigma_c^{129} \phi \mathcal{F}^{129}(\text{T}) \dots \dots \dots (7)$$

The isotopic ratios $\left[^{128}\text{Xe}/^{131}\text{Xe} \right]$ and $\left[^{130}\text{Xe}/^{131}\text{Xe} \right]$ are those measured for ^{238}Np fission. The absolute cumulative yields $Y(127)$, $Y(128)$, $Y(129)$, $Y(130)$, and $Y(131)$ are the values for ^{235}U fission taken from Table IX. The other terms appearing in these equations are the same as those defined for equations (4) and (5).

In the region of the krypton yields, the mass yield curve for ^{238}Np fission is somewhat different than that for ^{235}U fission. However, the error involved in the ^{82}Kr measurements is much larger

than the error involved in assuming $Y(82)/Y(83)$ to be the same for ^{238}Np fission as for ^{235}U fission. Thus, the fractional chain yield of ^{82}Br from ^{238}Np fission is:

$$F(^{82}\text{Br}) = \left[^{82}\text{Kr}/^{83}\text{Kr} \right] \left[Y(83)/Y(82) \right] - \left[Y(81)/Y(82) \right] \sigma_c^{81} \phi T/2 \dots (8)$$

Again, the isotope ratio $\left[^{82}\text{Kr}/^{83}\text{Kr} \right]$ is that measured for ^{238}Np fission, and the absolute cumulative yields $Y(81)$, $Y(82)$, and $Y(83)$ are the values for ^{235}U fission taken from Table IX.

Three ^{237}Np samples irradiated in the high flux (2×10^{14} neutrons/cm²/sec) were analyzed for ^{130}I , ^{128}I , and ^{82}Br fractional chain yields from ^{238}Np fission, as well as for cumulative yields.

The fractional chain yield results are shown in Table XVI.

Unfortunately, the HCl used to purify the neptunium contained a high level of natural bromine contamination. Thus it was possible only to set an upper limit on the ^{82}Br fractional chain yield, and to make no meaningful statement at all about the ^{80}Br yield. This upper limit was determined for sample A-1. The ratios $^{130}\text{Xe}/^{131}\text{Xe}$ and $^{128}\text{Xe}/^{131}\text{Xe}$ were obtained from samples A-2 and A-3. The observed ^{128}I fractional chain yields are exceptionally high -- being about half the ^{130}I fractional chain yields. The possibility of naturally occurring iodine in the HCl again cannot be ruled out. Thus the measured ^{128}I values can only be taken as upper limits on the ^{128}I fractional chain yield. The major portion of the error in the ^{130}I

yields is due to uncertainties in the average neutron flux. The average values of the various fractional chain yields are shown in the last column of Table XVI.

Summary of the Absolute Independent Yields

The absolute independent yields of the shielded bromine and iodine isotopes from the fission of ^{235}U , ^{233}U , and ^{239}Pu are summarized in Table XVII. Also shown for comparison are the previous mass spectrometric results obtained by Kennett (44) and Pleva (50) along with some radiochemical data on the ^{82}Br yields.

The present ^{130}I yield from the fission of ^{239}Pu agrees with Kennett's result, but the ^{130}I yields from the fission of ^{235}U and ^{233}U are somewhat lower than reported previously. In the case of ^{235}U fission, Pleva's results were obtained from a sample of a reactor fuel rod, which had been subjected to a very long irradiation. His measurements necessarily involved large neutron capture corrections, and consequently a greater margin of error. The present value, on the other hand, involves comparatively small neutron capture corrections.

The present ^{128}I yield from ^{235}U fission agrees with Pleva's value but is a factor of three lower than Kennett's value. The present ^{128}I yields from the fission of ^{233}U and ^{239}Pu are also a factor of three or four lower than Kennett's values. The latter results might have been affected by natural iodine contamination. It should be pointed out in this context that Kennett's data represent

TABLE XVII

SUMMARY OF ABSOLUTE INDEPENDENT YIELD DATA

Fission Product	Fissile Nuclide	Present Work	Kennett (44)	Pleva (50)	Radiochemical
⁸⁰ Br	²³⁵ U	$(6 \pm 1) \times 10^{-7}\%$	$4.0 \times 10^{-5}\%$	$4 \times 10^{-7}\%$	- - - - -
"	²³³ U	$< 2 \times 10^{-5}\%$	$2.34 \times 10^{-4}\%$	- - - - -	- - - - -
"	²³⁹ Pu	$< 1.5 \times 10^{-6}\%$	- - - - -	- - - - -	- - - - -
⁸² Br	²³⁵ U	$(4.6 \pm 0.5) \times 10^{-5}\%$	$4.5 \times 10^{-4}\%$	$5.4 \times 10^{-5}\%$	$3 \times 10^{-5}\%$ (43)
"	²³³ U	$(1.20 \pm 0.15) \times 10^{-3}\%$	$2.28 \times 10^{-3}\%$	- - - - -	$7.46 \times 10^{-4}\%$ (51)
"	²³⁹ Pu	$(1.52 \pm 0.18) \times 10^{-4}\%$	- - - - -	- - - - -	$3.6 \times 10^{-3}\%$ (52)
¹²⁶ I	²³⁵ U	$< 1 \times 10^{-6}\%$	- - - - -	- - - - -	- - - - -
¹²⁸ I	²³⁵ U	$(1.4 \pm 0.3) \times 10^{-5}\%$	$5.0 \times 10^{-5}\%$	$1.8 \times 10^{-5}\%$	- - - - -
"	²³³ U	$(3.3 \pm 0.5) \times 10^{-5}\%$	$1.34 \times 10^{-4}\%$	- - - - -	- - - - -
"	²³⁹ Pu	$(8.7 \pm 1.0) \times 10^{-5}\%$	$(2.15 \pm 0.05) \times 10^{-4}\%$	- - - - -	- - - - -
¹³⁰ I	²³⁵ U	$(2.3 \pm 0.3) \times 10^{-4}\%$	$5.11 \times 10^{-4}\%$	$4.7 \times 10^{-4}\%$	- - - - -
"	²³³ U	$(2.91 \pm 0.17) \times 10^{-3}\%$	$(3.9 \pm 0.3) \times 10^{-3}\%$	- - - - -	- - - - -
"	²³⁹ Pu	$(5.06 \pm 0.17) \times 10^{-3}\%$	$(5.2 \pm 0.4) \times 10^{-3}\%$	- - - - -	- - - - -

only one or two independent determinations of each yield, whereas the present data represent from three to five consistent measurements for each yield. Thus the possibility of serious error arising from contamination is greatly reduced in the present case.

The present ^{126}I yield from ^{235}U fission is only an upper limit. No previous attempts to measure this yield have been reported.

The present ^{82}Br and ^{80}Br yields from ^{235}U fission agree with Pleva's results within the limits of experimental error (within two standard deviations). The high yields that Kennett obtained for ^{82}Br and ^{80}Br appear to be the result of contamination. If Kennett's ^{80}Br yield from ^{233}U fission is assumed to be entirely due to atmospheric contamination, then an appropriate correction to his ^{82}Br yield brings it into agreement with the present ^{82}Br yield.

Most of the ^{82}Br yields reported in the literature are taken from radiochemical determinations. An early value for the ^{82}Br yield from ^{235}U fission was obtained by Feldman, Glendenin, and Edwards (43). It is of the same order of magnitude as the values obtained by mass spectrometry. The ^{82}Br yield measured radiochemically by Santry and Yaffe (51) for ^{233}U fission is about half the present value. Marsden and Yaffe (52) report a ^{82}Br yield from ^{239}Pu fission that is 20 times the mass spectrometric result. However, the mass spectrometric measurements of the independent yields of the shielded nuclides are believed to be more accurate than the radiochemical determinations, which involve detailed chemical separations and low counting rates. On the

other hand, the only serious source of error in the mass spectrometric determinations is natural contamination. The possibility of bromine or krypton contamination contributing significantly to the observed ^{82}Br yields has been ruled out in the present study by the low level of the observed ^{80}Kr abundances.

In summary, then, the present results for the independent yields of ^{82}Br , ^{128}I , and ^{130}I from the fission of ^{235}U , ^{233}U , and ^{239}Pu as well as the ^{80}Br yield from the fission of ^{235}U can be accepted with confidence. The yields of ^{80}Br from the fission of ^{233}U and ^{239}Pu and of ^{126}I from the fission of ^{235}U are to be taken as upper limits.

Summary of Fractional Chain Yields

The quantity of greatest interest to the theories of nuclear charge distribution in fission is the fractional chain yield of a given nuclide, i.e., the absolute independent yield of the nuclide divided by the cumulative yield of its mass chain. The average values of the fractional chain yields of ^{80}Br , ^{82}Br , ^{126}I , ^{128}I , and ^{130}I from each of the four fissile nuclides studied are summarized in Table XVIII. The fissile nuclides are arranged from left to right in the order of increasing charge density of the compound fissioning nucleus. The ^{238}Np yields are taken directly from Table XVI. The fractional chain yields from ^{235}U , ^{239}Pu , and ^{233}U are calculated

TABLE XVIII

SUMMARY OF FRACTIONAL CHAIN YIELD DATA

Fission Process	$^{238}\text{Np} + n$	$^{235}\text{U} + n$	$^{239}\text{Pu} + n$	$^{233}\text{U} + n$
Charge density of compound fissioning nucleus	0.3891	0.3898	0.3917	0.3932
$F(^{80}\text{Br})$	- - -	$(6 \pm 1) \times 10^{-6}$	$< 3 \times 10^{-5}$	$< 1 \times 10^{-4}$
$F(^{82}\text{Br})$	$\leq 1.4 \times 10^{-3}$	$(1.9 \pm 0.2) \times 10^{-4}$	$(9.5 \pm 1.1) \times 10^{-4}$	$(1.60 \pm 0.20) \times 10^{-3}$
$F(^{126}\text{I})$	- - -	$< 3 \times 10^{-5}$	- - -	- - -
$F(^{128}\text{I})$	$\leq (2.4 \pm 0.6) \times 10^{-4}$	$(3.7 \pm 0.8) \times 10^{-5}$	$(1.23 \pm 0.14) \times 10^{-4}$	$(3.0 \pm 0.5) \times 10^{-5}$
$F(^{130}\text{I})$	$(5.4 \pm 1.8) \times 10^{-4}$	$(1.15 \pm 0.15) \times 10^{-4}$	$(1.87 \pm 0.06) \times 10^{-3}$	$(1.12 \pm 0.07) \times 10^{-3}$

from the absolute independent yields of the present work and from the accepted values of the cumulative yields summarized in Table IX.

It is instructive to plot the fractional chain yield of a given fission product as a function of the charge density of the compound fissioning nucleus. This is done in Figure 18 for the ^{82}Br and ^{80}Br yields, and in Figure 19 for the ^{130}I and ^{128}I yields. It can be seen from Figure 18 that the logarithm of the ^{82}Br yields from ^{235}U , ^{239}Pu , and ^{233}U fission increases almost linearly with this charge density. This trend can be understood as a shifting of the most probable charge to progressively higher values as the charge density of the fissioning nucleus increases. The independent ^{82}Br and ^{80}Br yields lie on the high side of the charge distribution curve, and therefore increase in an approximately exponential manner. The ^{82}Br fractional chain yield from ^{238}Np does not conform to this trend. However, this last measurement represents an upper limit only. The upper limits of the ^{80}Br fractional chain yields are consistent with the general trend.

The ^{130}I and ^{128}I fractional chain yields in Figure 19 do not display this simple linear dependence on the charge density of the fissioning nucleus. We do observe an increase in the ^{130}I fractional chain yield in going from ^{235}U fission to ^{233}U fission, as we would expect. However, the ^{130}I yields from ^{238}Np and ^{239}Pu fission are too high. We would expect the ^{128}I yield from ^{235}U fission to be lower than that from ^{233}U fission, whereas, in fact, the two yields

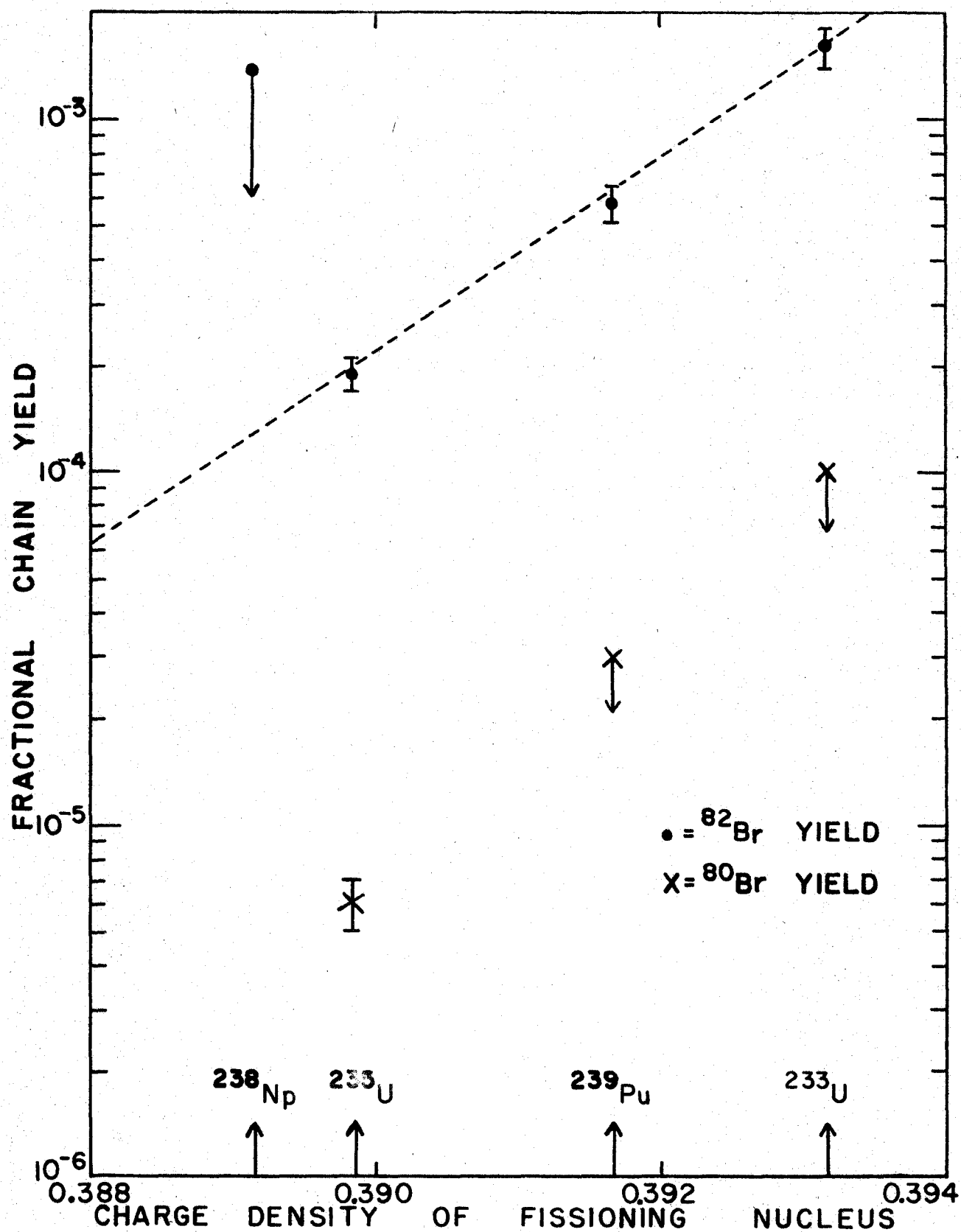


Fig. 18. ${}^{80}\text{Br}$ and ${}^{82}\text{Br}$ fractional chain yields as a function of the charge density of the compound fissioning nucleus.

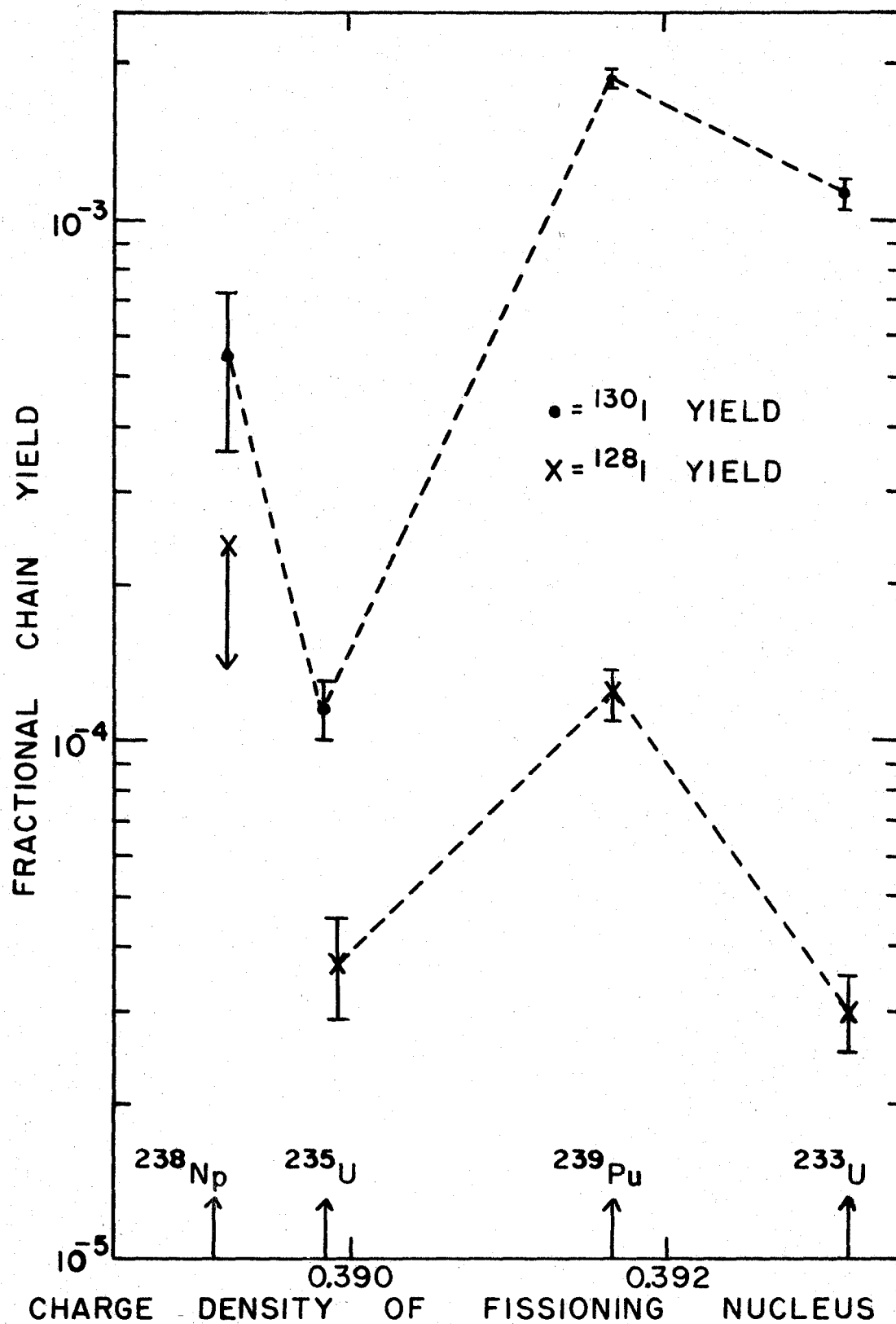


Fig. 19. ^{128}I and ^{130}I fractional chain yields as a function of the charge density of the compound fissioning nucleus.

are about equal. Furthermore, the ^{128}I yield from ^{239}Pu fission is considerably higher than the two yields from ^{235}U fission and ^{233}U fission. Thus there appears to be no simple correlation between the iodine fractional chain yields and the charge density of the fissioning nucleus.

Predictions by Conventional Charge Distribution Theories

In Table XIX the values of the fractional chain yields predicted by conventional theories of charge distribution in fission are compared with the experimental results. For each calculation, a Gaussian charge distribution of the following form is assumed:

$$P(Z) = \frac{1}{\sqrt{c\pi}} e^{-\frac{(Z - Z_P)^2}{c}} \dots\dots\dots (9)$$

The most probable charge is calculated by a number of approaches: equal charge displacement (ECD), maximum energy release (MER), and Wahl's empirical Z_p function. For the ECD and MER calculations, a constant value of $c = 1.00$ has been assumed. For the calculations from Wahl's empirical Z_p function, c has been allowed to vary with fission product mass number in order to obtain the best agreement with the experimental data.

In the bromine region the ECD calculations are in reasonable agreement with experiment for the ^{80}Br yield from the fission of ^{235}U and for the ^{82}Br yields from the fission of ^{235}U and ^{233}U . Also, the ECD values for the ^{82}Br yield from ^{238}Np fission and the

TABLE XIX

FRACTIONAL CHAIN YIELDS CALCULATED FROM CONVENTIONAL APPROACHES TO CHARGE DISTRIBUTION IN FISSION

Fission Product	Fissile Nuclide	Yield from ECD (C=1.00)	Yield Calculated from Wahl	Yield from MER (C=1.00)	Experimental Yield
^{80}Br	^{235}U	4.70×10^{-6}	5.81×10^{-6} (C=0.94)	- - -	$(6 \pm 1) \times 10^{-6}$
"	^{239}Pu	7.39×10^{-5}	- - -	- - -	$\leq 3 \times 10^{-5}$
"	^{233}U	2.77×10^{-5}	- - -	- - -	$\leq 1 \times 10^{-4}$
^{82}Br	^{238}Np	3.85×10^{-4}	- - -	- - -	$\leq 1.4 \times 10^{-3}$
"	^{235}U	2.22×10^{-4}	1.98×10^{-4} (C=0.78)	- - -	$(1.9 \pm 0.2) \times 10^{-4}$
"	^{239}Pu	2.05×10^{-3}	- - -	- - -	$(9.5 \pm 1.1) \times 10^{-4}$
"	^{233}U	2.05×10^{-3}	1.44×10^{-3} (C=0.78)	- - -	$(1.60 \pm 0.20) \times 10^{-3}$
^{128}I	^{235}U	5.51×10^{-7}	- - -	3.78×10^{-5}	$(3.7 \pm 0.8) \times 10^{-5}$
"	^{239}Pu	4.70×10^{-6}	- - -	6.96×10^{-5}	$(1.23 \pm 0.14) \times 10^{-4}$
"	^{233}U	1.05×10^{-5}	- - -	9.37×10^{-5}	$(3.0 \pm 0.5) \times 10^{-5}$
^{130}I	^{238}Np	7.85×10^{-5}	- - -	- - -	$(5.4 \pm 1.8) \times 10^{-4}$
"	^{235}U	1.05×10^{-4}	- - -	1.26×10^{-4}	$(1.15 \pm 0.15) \times 10^{-4}$
"	^{239}Pu	5.59×10^{-4}	- - -	2.22×10^{-4}	$(1.87 \pm 0.06) \times 10^{-3}$
"	^{233}U	1.04×10^{-3}	- - -	3.85×10^{-4}	$(1.12 \pm 0.07) \times 10^{-3}$

ECD = Postulate of equal charge displacement

MER = Postulate of maximum energy release (26).

Wahl = Wahl's empirical Z_p function (30).

^{80}Br yield from ^{233}U fission are within the upper limits obtained experimentally. The calculated ^{80}Br and ^{82}Br yields from ^{239}Pu fission are each high by a factor of two. However, the ECD calculations in the last cases are not as accurate, because the most stable charge, Z_A , on the complementary fragments is in the region of a proton shell.

The ^{80}Br yield from ^{235}U fission and the ^{82}Br yields from the fission of ^{235}U and ^{233}U have also been calculated from Wahl's empirical Z_p function. A value of $c = 0.94$ gives agreement for the 80 mass chain, and a value of 0.78 gives agreement for the 82 mass chain. Both these values are within the limits prescribed by Wahl for the value of c , i.e., $c = 0.86 \pm 0.15$ (31). Thus the independent bromine yield data are consistent with the general empirical trends observed by Wahl.

The ^{130}I yields calculated from the ECD postulate for the fission of ^{235}U and ^{233}U are in good agreement with the experimental results. However, the predicted ^{130}I yields for the fission of ^{238}Np and ^{239}Pu are far too low. The ^{128}I yields predicted by the ECD postulate are too low for all the fissile nuclides. The postulate of maximum energy release gives better agreement for the 128 mass chain.

However, no theory of charge distribution is able to explain the irregular variation of the ^{128}I and ^{130}I fractional chain yields with charge density of the fissioning nucleus, as illustrated in Figure 19. Every theory predicts an approximately linear increase in

Z_p with increasing charge density, and thus a corresponding increase in the logarithm of the fractional chain yields. Also, there is no reason to believe that the shape of the charge distribution curve changes drastically from one fissile nuclide to another. We are thus forced to the conclusion that the irregularities in the observed ^{128}I and ^{130}I fractional chain yields result from variations in the neutron emission probabilities from the prompt fission fragments. This situation is analogous to the fine structure in the mass yield curve.

Neutron Emission Considerations

In order to understand more clearly how such irregularities might arise from neutron emission effects, let us pursue a suggestion originally proposed by Cameron (53). He suggested that the observed independent yields of the shielded nuclides might be due in large part to multi-neutron emission from prompt fragments with the same charge, but with considerably higher mass numbers. For instance, the observed independent yield of ^{130}I can be written as follows:

$$Y(^{130}\text{I}) = P_0 y(^{130}\text{I}) + P_1 y(^{131}\text{I}) + P_2 y(^{132}\text{I}) + P_3 y(^{133}\text{I}) + \dots \quad (10)$$

where $y(^{130}\text{I})$ = prompt independent yield of ^{130}I , i.e., the independent yield before neutron emission,

P_0 = probability of emitting zero neutrons from ^{130}I ,

P_1 = probability of emitting one neutron from ^{131}I , etc.

The probabilities of multi-neutron emission decrease very rapidly as the number of neutrons increases. However, the prompt independent yields of the iodine fragments increase rapidly with mass number in the region of masses 128-136. It is possible that this increase in independent yield may offset to some degree the decrease in neutron emission probability. In such a case a considerable portion of the observed ^{130}I independent yield might actually result from iodine fragments formed at mass 132 or 133.

The probabilities for emitting various numbers of neutrons from a given prompt fission fragment depends upon the amount of excitation energy allotted to that fragment by the particular fission reaction that produced it. If more excitation energy were allotted to the iodine fragments from the fission of ^{239}Pu and ^{238}Np than to those from the fission of ^{235}U and ^{233}U , then the neutron emission probabilities from higher mass iodine fragments would be considerably enhanced. This would explain the high observed yields of ^{128}I and ^{130}I from the fission of ^{239}Pu and ^{238}Np .

There is reason to believe that additional excitation energy is, in fact, given to the iodine fragments from the fission of ^{239}Pu and ^{238}Np . First of all, the average total number of neutrons emitted during the fission of ^{239}Pu is 2.88, whereas for ^{235}U and ^{233}U the number is only about 2.50. This indicates that the excitation energies given to the ^{239}Pu fission fragments are higher, on the average, than those given to the ^{235}U or ^{233}U fission fragments.

Second, the semi-empirical mass law predicts greater energy releases in the formation of the iodine fragments from the fission of both ^{238}Np and ^{239}Pu . Table XX shows the total amount of energy released in the formation of various iodine isotopes and their complementary fragments from the four fission processes studied. The energies are calculated from Seeger's semi-empirical mass tables (54). In the case of ^{239}Pu fission, there is from 13 to 15 Mev more energy released for each iodine isotope than for ^{235}U fission, and about 10 Mev more than for ^{233}U fission. If even a small fraction of this increase appeared as additional excitation energy on the iodine fragments, then the neutron emission probabilities from these fragments would be considerably enhanced for ^{239}Pu fission. The energy releases for ^{238}Np fission are also somewhat larger than for the fission of the two uranium isotopes.

To place the foregoing considerations on a more quantitative basis, let us now proceed to calculate the fractional chain yields of ^{128}I and ^{130}I for the various fission processes by making use of equation 10. To begin with, the prompt independent yield of a given nuclide is the product of the prompt mass yield at the particular mass number and the prompt fractional chain yield for the particular nuclear charge. Prompt mass yield data for the fission of ^{235}U , ^{233}U , and ^{239}Pu are taken from Milton and Fraser (20). The prompt mass yields for ^{238}Np fission are assumed to be the same as for ^{235}U fission in the region of mass 128 to 133. It was seen in the previous section that there are close similarities in the cumulative yield curves for

TABLE XX

TOTAL ENERGY RELEASES FROM THE FOUR FISSION PROCESSES STUDIED

Fission Fragments	$^{235}\text{U} + \text{n}$	$^{233}\text{U} + \text{n}$	$^{238}\text{Np} + \text{n}$	$^{239}\text{Pu} + \text{n}$
^{128}I + complement	173.33	178.09	180.52	188.55
^{129}I + complement	179.75	184.18	186.84	194.44
^{130}I + complement	181.35	185.42	188.38	195.59
^{131}I + complement	187.05	190.76	194.01	200.79
^{132}I + complement	187.91	191.22	194.83	201.21
^{133}I + complement	192.85	195.75	199.72	205.68

All energies are in Mev. Calculations are based on Seeger's semi-empirical mass tables (54).

these two nuclides in this mass region.

The prompt fractional chain yields are determined from a universal Gaussian charge distribution curve as given in equation 9. The width, c , of the curve is taken to be constant at 1.00. The most probable charge at each mass number is calculated from the simplest theory that gives reasonable agreement with experimental data in general -- the postulate of equal charge displacement.

In order to calculate the neutron emission probabilities from the various fragments, it is necessary to know the neutron binding energies of the fragments and the amount of excitation energy given to the fragments. The neutron binding energies are taken from Seeger's semi-empirical mass tables (54). The average excitation energy per fragment is assumed to be a linear function of fragment mass, i.e., to be of the following form:

$$E_x(A) = K(A-A_0) \dots\dots\dots (11)$$

This form is analogous to Terrell's universal expression for the number of prompt neutrons emitted per fragment (21). By using such a linear excitation energy relationship, Thind and Tomlinson (55) have been able to reproduce quite well the observed neutron emission curve, including "fine structure", for the heavy mass peak from ^{235}U fission.

The average excitation energy, given by equation 11 for a particular mass number, is assumed to be independent of nuclear charge.

In other words the iodine fragments of interest, having been formed by a less probable division of nuclear charge, receive the same excitation energies as the fragments formed by the most probable division of charge. This assumption receives support from recent data by Gordon et al. (56) on the range of the shielded nuclide ^{136}Cs . They found that the kinetic energy release for ^{136}Cs and its complementary fragment is considerably less than the average for the 136 mass chain. This kinetic energy deficit is about equal to the decrease in the calculated total energy release for ^{136}Cs and its complementary fragment, in comparison with the total energy release for the most probable fragments at mass 136. Since the excitation energy is the difference between the total energy release and the kinetic energy, these authors conclude that the excitation energy is the same on all fragments at mass 136.

For the fission of ^{235}U and ^{233}U , the values of the constants K and A_0 in equation 11 are those used by Thind and Tomlinson (55) to reproduce the observed neutron emission curve for ^{235}U fission. To explain the higher independent yields for ^{239}Pu and ^{238}Np fission, the average excitation energies must be shifted to higher values. This is accomplished by keeping the slope, K , of the excitation energy line constant, but allowing A_0 to vary. It has already been shown that additional excitation energy is likely on the fission fragments from ^{239}Pu and ^{238}Np fission.

Some assumptions must be made regarding the distribution of

excitation energies about the average value at a given mass number. This is particularly important in the mass region of interest, i.e., below mass 134. Here the average excitation energy is less than the binding energy of the first neutron. If there were no distribution in excitation energies, then the number of neutrons emitted per fragment would fall abruptly from one to zero as the mass decreased below 134. Such an abrupt change is not supported by experimental data. The number of neutrons emitted actually decreases gradually to zero as the mass approaches 126. Since there are virtually no data on the distribution of excitation energies of a particular fragment, the simplest possible assumptions are made. The distributions are assumed to be Gaussian, with a constant width. The standard deviation of these distributions must be of the same order of magnitude as the average excitation energies, i.e., it must be at least several Mev. Energy conservation considerations would restrict its value to less than 10 Mev. Within this range, the standard deviation is treated as a variable parameter to be fitted to the experimental results of the present work.

A method given by Jackson (57) has been used to calculate the neutron emission probabilities from the neutron binding energies and the excitation energies allotted to the fragments. This method is now outlined briefly: As the excitation energy increases beyond the binding energy of the first neutron, the emission probability for one neutron rises from zero to one in a step function. The

situation becomes more complicated as the excitation energy passes the binding energy of the second neutron. The fragment may either emit two neutrons with low kinetic energies or one neutron with a high kinetic energy. In order to derive the neutron emission probabilities for more than one neutron, a neutron energy spectrum of the form: $n(\epsilon) \propto e^{-\epsilon/T}$ is assumed. ϵ is the kinetic energy of the neutron and T is the nuclear temperature. The probability that exactly n neutrons will be emitted at excitation energy E_x is then given by the neutron evaporation function:

$$P(E_x, n) = I(\Delta_n, 2n - 3) - I(\Delta_{n+1}, 2n - 1) \dots \dots \dots (12)$$

where

$$\Delta_n = \frac{E_x - \sum_{i=1}^n B_i}{T}$$

B_i = binding energy of the i 'th neutron,

T = nuclear temperature, taken to be 1.4 Mev for these calculations,

$I(u, p)$ = Incomplete gamma function. Values may be found in the tables by Pearson (58). The neutron evaporation functions for ^{131}I are shown in Figure 20.

The excitation energies for a given fragment are actually distributed about an average value. The probability of emitting n neutrons from this fragment is given by the overlap integral of the excitation energy distribution and the neutron evaporation function, $P(E_x, n)$.

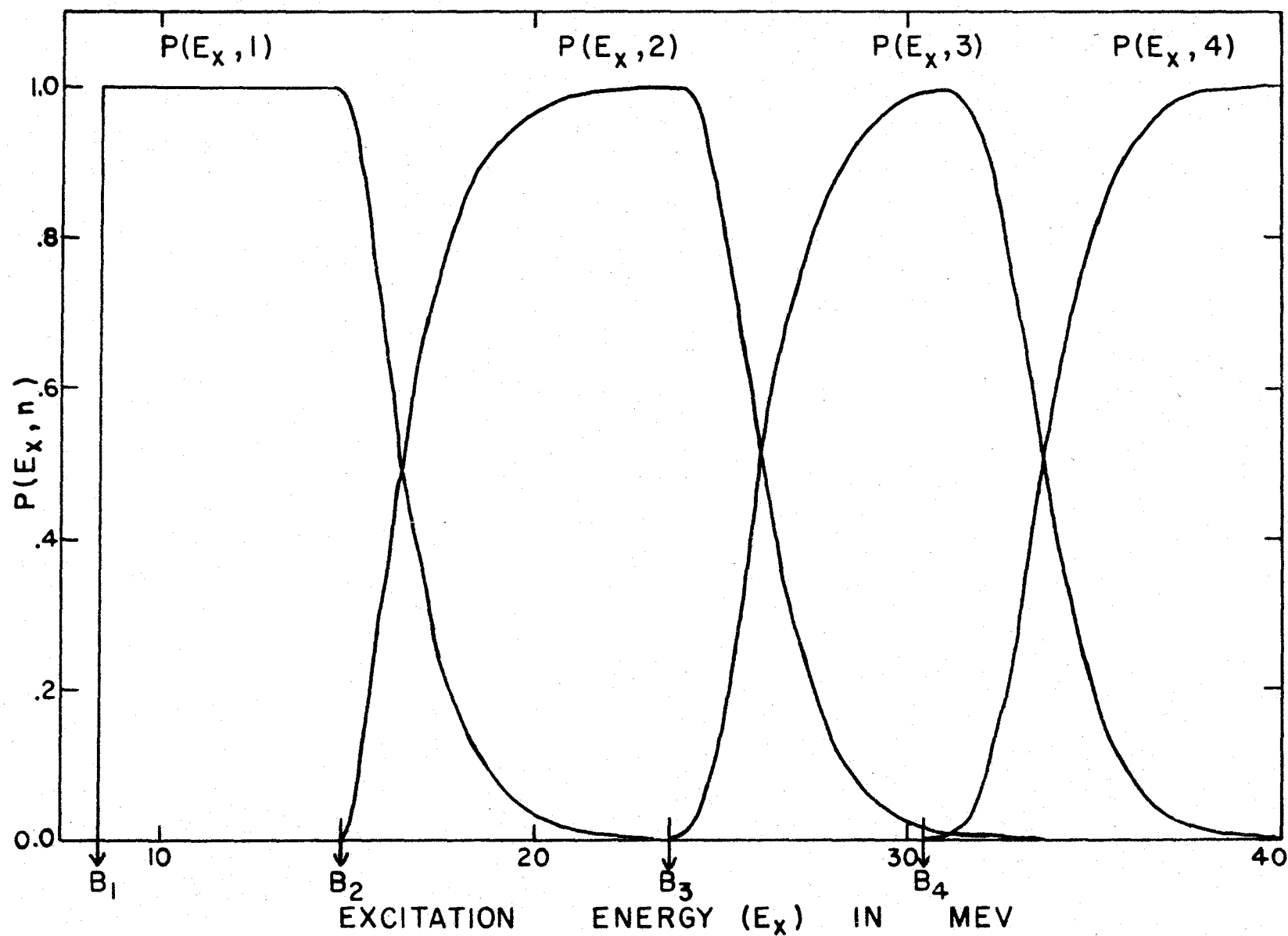


Fig. 20. Calculations of the neutron evaporation function, $P(E_x, n)$ for ^{131}I . B_i = binding energy of the i 'th neutron.

Let us summarize briefly the assumptions made in carrying out these calculations:

- (1) prompt mass yield data as given by Milton and Fraser,
- (2) the most probable charge on the prompt fragments given by the postulate of equal charge displacement,
- (3) a smooth prompt charge distribution curve with a Gaussian shape and a constant width,
- (4) a linear dependence of mean excitation energy per fragment upon fragment mass,
- (5) the mean excitation energy per fragment independent of nuclear charge at a given mass number,
- (6) a distribution of excitation energies about the mean with a Gaussian shape and a constant width,
- (7) the neutron evaporation theory of Jackson.

The results of these calculations are shown in Table XXI where they are compared with the experimental results. Three different values have been used for the standard deviation of the excitation energy distributions, i.e., $\sigma_x^- = 4.0, 5.0, \text{ and } 6.0 \text{ Mev.}$ For each value the parameter A_0 for ^{239}Pu and ^{238}Np fission has been adjusted to give the best fit to experimental data for these two fissile nuclides.

All in all, a value of $\sigma_x^- = 5.0 \text{ Mev}$ gives reasonable agreement with most of the experimental data, at least to within a factor of two. One could not really hope for more exact agreement

TABLE XXI

FRACTIONAL CHAIN YIELDS CALCULATED FROM NEUTRON EMISSION MECHANISM

Fission Product	Fissile Nuclide	Calculated fractional chain yields			Experimental fractional chain yield
		$\sigma_x^- = 4.0 \text{ Mev}$	$\sigma_x^- = 5.0 \text{ Mev}$	$\sigma_x^- = 6.0 \text{ Mev}$	
^{128}I	^{235}U	0.979×10^{-6}	1.96×10^{-6}	3.97×10^{-6}	$(3.7 \pm 0.8) \times 10^{-5}$
"	^{239}Pu	6.27×10^{-5}	6.17×10^{-5}	6.73×10^{-5}	$(1.23 \pm 0.14) \times 10^{-4}$
"	^{233}U	0.702×10^{-5}	1.22×10^{-5}	2.16×10^{-5}	$(3.0 \pm 0.5) \times 10^{-5}$
^{130}I	^{238}Np	2.59×10^{-4}	2.35×10^{-4}	2.30×10^{-4}	$(5.4 \pm 1.8) \times 10^{-4}$
"	^{235}U	0.965×10^{-4}	1.58×10^{-4}	2.58×10^{-4}	$(1.15 \pm 0.15) \times 10^{-4}$
"	^{239}Pu	3.16×10^{-3}	2.88×10^{-3}	2.80×10^{-3}	$(1.87 \pm 0.06) \times 10^{-3}$
"	^{233}U	0.769×10^{-3}	1.12×10^{-3}	1.61×10^{-3}	$(1.12 \pm 0.07) \times 10^{-3}$
Ao for ^{239}Pu and ^{238}Np		112.5	114.3	116.0	

Average excitation energy $\overline{E}_x(A) = K(A - A_0)$

For ^{235}U and ^{233}U fission, $K = 0.73$
 $A_0 = 123.8$

For ^{238}Np and ^{239}Pu fission, $K = 0.73$
 A_0 is fitted for each σ_x^- .

σ_x^- = standard deviation of the distributions in excitation energy.

than this on the basis of the simple assumptions made about the charge distribution on the prompt fragments and about the distribution in excitation energies. The only really serious discrepancy is the calculated ^{128}I yield from ^{235}U fission which is an order of magnitude too low. Quite possibly, the 50-proton shell does influence the most probable charge on the prompt fragments leading to the formation of ^{128}I in this case. Such an effect is predicted by the postulate of maximum energy release, but has not been considered in the equal charge displacement calculations leading to Table XXI.

A value of $\sigma_x^- = 6.0$ Mev gives somewhat better agreement for the ^{128}I yields, but makes the ^{130}I yields from ^{235}U and ^{233}U fission too high. A value of $\sigma_x^- = 4.0$ Mev is obviously too low.

A comparison of Tables XIX and XXI shows that the calculations based on neutron emission effects, particularly with $\sigma_x^- = 5.0$ Mev, are much more successful in accounting for the irregularities in the observed yields, than are the calculations based on conventional theories of charge distribution. In particular, the abnormally high yields of ^{128}I and ^{130}I from ^{239}Pu fission and of ^{130}I from ^{238}Np fission are explained.

In conclusion, a mass spectrometer has been used to obtain accurate absolute independent yield data on four shielded nuclides produced in fission. The observed fractional chain yields of ^{128}I and ^{130}I show irregularities that cannot be accounted for by conventional theories of charge distribution in fission. However,

these irregularities have been explained by calculations based on the details of prompt neutron emission. The observed fractional chain yields in other regions of the mass yield curve may also be affected by fluctuations in prompt neutron emission probabilities. As more independent yield data of high accuracy become available, it will be possible to subject this neutron emission mechanism to further tests.

APPENDIX A

RATIO OF THE NUMBER OF ^{238}Np TO ^{237}Np FISSIONS

We shall now derive equation (1) on page 47. During the irradiation of ^{237}Np the rate of change of the number of ^{238}Np atoms is given by the following differential equation:

$$\frac{dN(238)}{dt} = N(237)\sigma_c^{237} \phi - \lambda N(238)$$

$N(237)$ = number of ^{237}Np atoms. For the irradiation times of interest in this work, the number of ^{237}Np atoms can be taken as a constant throughout the irradiation period.

σ_c^{237} = ^{237}Np thermal neutron capture cross section.

σ_f^{238} = ^{238}Np thermal neutron fission cross section.

ϕ = thermal neutron flux

λ = effective decay constant of ^{238}Np in a reactor flux
= true decay constant + $\sigma_f^{238} \phi$

t = time from the beginning of the irradiation.

Using the boundary condition that $N(238) = 0$ when $t = 0$, we obtain the following solution to the differential equation:

$$N(238) = \frac{N(237) \sigma_c^{237} \phi}{\lambda} \left\{ 1 - \exp(-\lambda t) \right\}$$

The number of ^{238}Np fissions occurring in a time interval dt is

$$dN_{238}^f = \sigma_f^{238} \phi N(238) dt.$$

If the total irradiation is T , then the total number of ^{238}Np fissions is:

$$\begin{aligned} N_{238}^f &= \int_0^T \sigma_f^{238} \phi N(238) dt \\ &= \frac{N(237) \sigma_f^{238} \sigma_c^{237}}{\lambda} \phi^2 T \left\{ 1 - \frac{1 - \exp(-\lambda T)}{\lambda T} \right\} \end{aligned}$$

The number of ^{237}Np fissions for a total irradiation time T is simply:

$$N_{237}^f = N(237) \sigma_f^{237} \phi T$$

where $\sigma_f^{237} = ^{237}\text{Np}$ thermal neutron fission cross section.

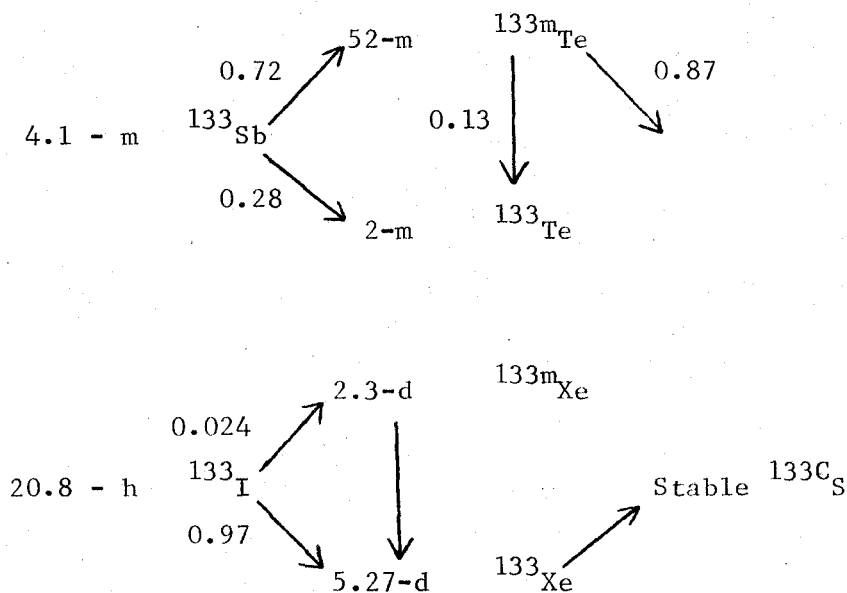
Thus the ratio of the number of ^{238}Np fissions to that of ^{237}Np fissions is:

$$\frac{N_{238}^f}{N_{237}^f} = \frac{\sigma_f^{238} \sigma_c^{237}}{\sigma_f^{237} \lambda} \phi \left\{ 1 - \frac{1 - \exp(-\lambda T)}{\lambda T} \right\}$$

APPENDIX B

CORRECTIONS FOR THE PARTIAL DECAY OF UNSTABLE XENON AND KRYPTON ISOTOPES

We shall derive the expression for the partial decay of ^{133}Xe produced in the fission of ^{238}Np . The correction expressions for the other unstable isotopes are quite similar. To begin with, the mass 133 chain appears as follows:



However, let us ignore the half-lives of short-lived chain members, and assume that the entire chain is formed initially as ^{133}I .

Furthermore, let us assume that the ^{133}I decays entirely to the ground state of ^{133}Xe , i.e., we are ignoring the 2.3-day isomer of ^{133}Xe .

Now if $N(^{133}\text{I})$ is the number of ^{133}I atoms in the sample at a time

t after irradiation begins, then

$$\frac{dN(^{133}\text{I})}{dt} = \left\{ ^{238}\text{Np fission rate} \right\} Y(133) - \alpha N(^{133}\text{I})$$

where $Y(133)$ = the cumulative yield of the mass 133 chain,

and α = decay constant of ^{133}I . From Appendix A, the ^{238}Np fission rate is:

$$\begin{aligned} & \frac{N(237) \sigma_f^{238} \sigma_c^{237} \phi^2}{\lambda} \left\{ 1 - \exp(-\lambda t) \right\} \\ & = C \left\{ 1 - \exp(-\lambda t) \right\} \text{ for an abbreviation.} \end{aligned}$$

Using the boundary condition that $N(^{133}\text{I}) = 0$ at $t = 0$, we obtain the following solution to the differential equation:

$$N(^{133}\text{I}) = C Y(133) \left[\frac{1 - \exp(-\alpha t)}{\alpha} - \frac{\exp(-\lambda t) - \exp(-\alpha t)}{\alpha - \lambda} \right]$$

The equation for the rate of change of the number of ^{133}Xe atoms during the irradiation is

$$\frac{d N(^{133}\text{Xe})}{dt} = \alpha N(^{133}\text{I}) - \beta N(^{133}\text{Xe})$$

where β = decay constant of ^{133}Xe . Thus, with the condition $N(^{133}\text{Xe}) = 0$ at $t = 0$, we obtain

$$\begin{aligned} N(^{133}\text{Xe}) &= C Y(133) \left[\frac{1 - \exp(-\beta t)}{\beta} \right. \\ &+ \frac{\lambda}{\alpha - \lambda} \frac{\exp(-\beta t) - \exp(-\alpha t)}{\alpha - \beta} - \frac{\alpha}{\alpha - \lambda} \frac{\exp(-\beta t) - \exp(-\lambda t)}{\lambda - \beta} \left. \right] \end{aligned}$$

Between the end of irradiation and the time of extraction of the rare gases, the differential equation for the number of ^{133}I atoms is simply

$$\frac{d N(^{133}\text{I})}{dt} = -\alpha N(^{133}\text{I}),$$

whereas the equation for the number of ^{133}Xe atoms remains the same as that used during the irradiation.

After extraction of the rare gases, the decay of ^{133}I no longer contributes to the abundance of ^{133}Xe . During this time the ^{133}Xe decays away in a simple exponential manner, i.e.,

$$\frac{d N(^{133}\text{Xe})}{dt} = -\beta N(^{133}\text{Xe})$$

Thus, if T is the total irradiation time, t' the time from the end of irradiation to the extraction of the rare gases, and T' the time from extraction to analysis, then at the time of analysis, the number of ^{133}Xe atoms is given by:

$$\begin{aligned} N(^{133}\text{Xe}) = & C Y(133) \left[\left\{ \frac{1 - \exp(-\beta T)}{\beta} + \frac{\lambda}{\alpha - \lambda} \frac{\exp(-\beta T) - \exp(-\alpha T)}{\alpha - \beta} \right. \right. \\ & \left. \left. - \frac{\alpha}{\alpha - \lambda} \frac{\exp(-\beta T) - \exp(-\lambda T)}{\lambda - \beta} \right\} \exp(-\beta t') \right. \\ & \left. + \left\{ 1 - \exp(-\alpha T) - \frac{\lambda}{\alpha - \lambda} (\exp(-\lambda T) - \exp(-\alpha T)) \right\} \right. \\ & \left. \times \frac{\exp(-\beta t') - \exp(-\alpha t')}{\alpha - \beta} \right] \exp(-\beta T) \end{aligned}$$

Now the 134 mass chain is assumed to be formed initially as stable ^{134}Xe . Thus, at the end of an irradiation time T , the total number of ^{134}Xe atoms is

$$\begin{aligned} N(^{134}\text{Xe}) &= \left\{ \text{total number of } ^{238}\text{Np fissions} \right\} Y(134) \\ &= C T \left(1 - \frac{1 - \exp(-\lambda T)}{\lambda T} \right) Y(134) \end{aligned}$$

where $Y(134) =$ cumulative yield at mass 134 for ^{238}Np fission.

Dividing the expression for $N(^{133}\text{Xe})$ by the expression for $N(^{134}\text{Xe})$ and solving for the ratio $Y(133)/Y(134)$, we obtain

$$\begin{aligned} \frac{Y(133)}{Y(134)} &= \left\{ \frac{N(^{134}\text{Xe})}{N(^{133}\text{Xe})} \right\} \times \\ &\exp(\beta T') T \left\{ 1 - \frac{1 - \exp(-\lambda T)}{\lambda T} \right\} \div \\ &\left[\left\{ \frac{1 - \exp(-\beta T)}{\beta} + \frac{\lambda}{\alpha - \lambda} \frac{\exp(-\beta T) - \exp(-\alpha T)}{\alpha - \beta} \right. \right. \\ &- \frac{\alpha}{\alpha - \lambda} \frac{\exp(-\beta T) - \exp(-\lambda T)}{\lambda - \beta} \left. \right\} \exp(-\beta t') \\ &+ \left. \left\{ 1 - \exp(-\alpha T) - \frac{\alpha}{\alpha - \lambda} \left(\exp(-\lambda T) - \exp(-\alpha T) \right) \right\} \right] \\ &\times \frac{\exp(-\beta t') - \exp(-\alpha t')}{\alpha - \beta} \end{aligned}$$

The ratio $N(^{134}\text{Xe})/N(^{133}\text{Xe})$ is measured directly by the mass spectrometer.

The various times — T , t' , T' — are carefully noted during the

experiments, and the decay constants are obtained from the literature.

Thus the relative yield $Y(133)/Y(134)$ can be calculated.

The expression for $Y(135)/Y(134)$ has the same form as the one derived above. As before, the entire mass 135 chain is assumed to be formed at ^{135}I . However, since a considerable fraction of the ^{135}Xe captures neutrons to form ^{136}Xe , the decay constant for ^{135}Xe must be modified. Thus, during irradiation, the effective decay constant for ^{135}Xe is:

$$\delta = \beta + \sigma_c^{135} \phi$$

where β = true decay constant of ^{135}Xe

σ_c^{135} = neutron capture cross section of ^{135}Xe

ϕ = neutron flux

The expressions for $Y(87)/Y(86)$ and $Y(88)/Y(86)$ are somewhat simpler, because the half-lives of the bromine precursors can be neglected. We thus have,

$$\frac{Y(87)}{Y(86)} = \left\{ \frac{N(^{86}\text{Kr})}{N(^{87}\text{Kr})} \right\} \exp \left\{ \epsilon(t' + T') \right\} T \left\{ 1 - \frac{1 - \exp(-\lambda T)}{\lambda T} \right\}$$

$$\div \left\{ \frac{1 - \exp(-\epsilon T)}{\epsilon} - \frac{\exp(-\lambda T) - \exp(-\epsilon T)}{\epsilon - \lambda} \right\}$$

where ϵ = decay constant of ^{87}Kr and $\left\{ N(^{86}\text{Kr})/N(^{87}\text{Kr}) \right\}$ is the isotopic ratio measured by the mass spectrometer.

The expression for $Y(88)/Y(86)$ is identical in form to the above expression.

APPENDIX C

NEUTRON CAPTURE CORRECTION TO THE OBSERVED ^{136}Xe YIELDS

In thermal neutron fission, ^{136}Xe is produced as a direct result of the decay of the mass 136 chain and also as a result of neutron capture on ^{135}Xe . The number of atoms of ^{136}Xe produced in a time interval dt during the irradiation of ^{238}Np is given by

$$dN(^{136}\text{Xe}) = Y(136) \left\{ ^{238}\text{Np fission rate} \right\} dt + \sigma_c^{135} \phi N(^{135}\text{Xe}) dt$$

where $Y(136)$ = true cumulative yield of the mass 136 chain

σ_c^{135} = neutron capture cross section of ^{135}Xe

ϕ = neutron flux

$N(^{135}\text{Xe})$ = number of ^{135}Xe atoms present at the given time. The

form of this expression has been discussed in Appendix B.

$$\left\{ ^{238}\text{Np fission rate} \right\} = C \left\{ 1 - \exp(-\lambda t) \right\} \quad \text{from Appendix B.}$$

Thus for a total irradiation time T , we have,

$$N(^{136}\text{Xe}) = Y(136) \int_0^T C \left\{ 1 - \exp(-\lambda t) \right\} dt + \sigma_c^{135} \phi \int_0^T N(^{135}\text{Xe}) dt$$

If the yield of ^{136}Xe is to be measured relative to ^{132}Xe , then we must also know the total number of ^{132}Xe atoms produced during irradiation.

$$\text{Thus } N(^{132}\text{Xe}) = Y(132) \int_0^T C \{1 - \exp(-\lambda t)\} dt$$

where $Y(132)$ = cumulative yield at mass 132.

We thus obtain the following expression for the true yield at mass 136 relative to that at mass 132:

$$\frac{Y(136)}{Y(132)} = \left\{ \frac{N(^{136}\text{Xe})}{N(^{132}\text{Xe})} \right\} - \frac{Y(135)}{Y(132)} \sigma_c^{-135} \phi$$

$$\times \left[\frac{1 - \frac{1 - \exp(-\delta T)}{\delta T}}{\delta} + \frac{\lambda}{\alpha - \lambda} \frac{\frac{1 - \exp(-\alpha T)}{\alpha T} - \frac{1 - \exp(-\delta T)}{\delta T}}{\delta - \alpha} \right. \\ \left. - \frac{\alpha}{\alpha - \lambda} \frac{\frac{1 - \exp(-\lambda T)}{\lambda T} - \frac{1 - \exp(-\delta T)}{\delta T}}{\delta - \lambda} \right]$$

$$\div \left[1 - \left\{ \frac{1 - \exp(-\lambda T)}{\lambda T} \right\} \right]$$

$\left\{ N(^{136}\text{Xe}) / N(^{132}\text{Xe}) \right\}$ is the isotopic ratio measured by means of the mass spectrometer.

$Y(135)$ = cumulative yield at mass 135

δ = effective decay constant of ^{135}Xe during irradiation
 = (true decay constant) + $\sigma_c^{-135} \phi$

α = decay constant of ^{135}I

λ = effective decay constant of ^{238}Np
 = (true decay constant) + $\sigma_f^{-238} \phi$.

APPENDIX D

NEUTRON CAPTURE CORRECTIONS IN THE INDEPENDENT YIELD DETERMINATIONS

In equations (4) and (5) on page 87, reference was made to the functions $f^{127}(T)$ and $f^{129}(T)$, which express the time dependence of the decay of the precursors of ^{127}I and ^{129}I respectively. These functions are derived from the data in Figure 17. Both functions have the same form; only the values of the respective constants are different. In each case, the entire chain yield is assumed to be initially formed at antimony.

The relevant differential equations are:

$$\frac{dN(\text{Sb})}{dt} = N_0 \sigma_f \phi Y - \alpha N(\text{Sb})$$

$$\frac{dN(\text{Te-isomer})}{dt} = p \alpha N(\text{Sb}) - \beta N(\text{Te-isomer})$$

$$\frac{dN(\text{Te-ground state})}{dt} = q \alpha N(\text{Sb}) + \beta N(\text{Te-isomer}) - \gamma N(\text{Te-ground state})$$

$$\frac{dN(\text{I})}{dt} = \gamma N(\text{Te-ground state})$$

$N(X)$ = number of atoms of species X present at a given time,

N_0 = number of nuclei of the fissioning species,

- σ_f^- = neutron fission cross section of the fissioning species,
 ϕ = neutron flux,
 Y = cumulative yield of the 127 or 129 chain,
 t = time from the beginning of the irradiation,
 α = decay constant of the antimony,
 β = decay constant of the isomeric state of tellurium,
 γ = decay constant of the ground state of tellurium,
 p = fraction of the antimony which decays to the isomeric state of tellurium,
 q = fraction of the antimony which decays to the ground state of tellurium.
 $p + q = 1$

The differential equations are solved with the boundary conditions that $N(X) = 0$ at $t = 0$ for every species X .

The neutron capture contribution to the total number of ^{128}I or ^{130}I atoms is then given by

$$\sigma_c^- \phi \int_0^T N(I) dt$$

where σ_c^- = neutron capture cross section of ^{127}I or ^{129}I ,

T = total irradiation time,

$N(I)$ = number of atoms of ^{127}I or ^{129}I present at a given time.

The expression for $\mathcal{J}(T)$ is then:

$$\begin{aligned} \mathcal{J}(T) = & \frac{T}{2} - \frac{1 - \frac{1 - \exp(-\gamma T)}{\gamma T}}{\gamma} \\ & - \frac{\gamma}{\gamma - \alpha} \left(q - \frac{p\beta}{\alpha - \beta} \right) \left[\frac{1 - \frac{1 - \exp(-\alpha T)}{\alpha T}}{\alpha} - \frac{1 - \frac{1 - \exp(-\gamma T)}{\gamma T}}{\gamma} \right] \\ & - \frac{p\alpha\gamma}{(\gamma - \beta)(\alpha - \beta)} \left[\frac{1 - \frac{1 - \exp(-\beta T)}{\beta T}}{\beta} - \frac{1 - \frac{1 - \exp(-\gamma T)}{\gamma T}}{\gamma} \right] \end{aligned}$$

In the case of the neutron capture correction to ^{82}Br , it can be assumed that the mass 81 chain is formed initially at stable ^{81}Br .

In this case, we have simply $\mathcal{J}(T) = \frac{T}{2}$

This is the expression used in equation (3) on page 87.

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